ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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TECHNICAL PROGRESS REVIEWS

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ISOTOPES AND RADIATION TECHNOLOGY

A REVIEW OF RECENT DEVELOPMENTS

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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VOLUME 1

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Foreword

Isotopes and Radiation Technology is a Technical Progress Review prepared by the Isotopes Development Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

P. S. BAKER, ROBERT H. LAFFERTY, Jr.,
MARTHA GERRARD, and D. A. FUCCILLO, Editors
A. F. RUPP, Advisory Editor
Oak Ridge National Laboratory

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Section

I

Isotopes and Radiation Technology

Isotope Technology Development

Some new uses of radioactive isotopes as tracers in industrial and other large-scale engineering applications were described in the Spring 1964 issue of *Isotopes and Radiation Technology*. In the present issue the discussion of tracers is continued with a description of several recent applications of radioactive tracers to analytical chemistry and new developments in counting equipment and techniques.

Tracers in Analysis

Radiotracers are quite useful in solving analytical problems. In batch testing of process streams, for example, the extreme sensitivity of the tracer method eliminates the necessity in many cases for lengthy quantitative separation procedures. In other problems, radioactive tracer techniques generally are marked by straightforward, rapid analysis and simpler equipment than in conventional approaches. Two studies have recently been reported which explore the uses of radiotracers in analytical chemistry and point to areas needing further work.

Reynolds and Leddicotte of Oak Ridge National Laboratory (ORNL) summarized, with 767 references, the uses of radioactive tracers in analytical chemistry.[1] The paper results from a literature search done as part of an ORNL study (supported by the AEC Division of Isotopes Development) intended to locate the gaps in radioisotope methodology. The coverage extended through 1961, with isolated references to reports in 1962. In addition to discussing the preparation of radiotracers and techniques, the authors describe the major fields of analytical applications: (1) isotope dilution, (2) radioreagents, (3) radiometric methods, and (4) studies of methods. There are two extensive tables in that review, parts of which are reproduced here in Tables I-1 and I-2 as examples. No attempt was made in the tables to refer to every publication in the field, and, in particular, only typical examples of the organic chemistry literature are given.

Driscoll et al.,[2] in exploring radiometric methods for industrial process control, surveyed the various beta-emitting isotopes that have possible application in radiometric analysis. In such a method the radioactivity of a reagent is standardized in terms of chemical equivalence, and an excess of the reagent is added to a solution. The ratio of the excess activity to the total activity then is used to determine one of the reactants. The technique used in the analyses reported by them was to measure the radioactivity of the excess reagent, exclusively, in a single step. They found that about one-third of the elements of the periodic table can be determined by use of this technique and noted reagents that may be used for the ions sought (Table I-3).

Table I-1 INORGANIC APPLICATIONS OF RADIOTRACERS

Element and tracer	Method*	Remarks	References†
Aluminum			
^{45}Ca	RR	Sorption on ^{45}Ca-loaded resin	7A
^{32}P	RR	Pptn. as phosphate	51B, 9T
^{131}I	RR, ID	Hydrol., reac. with I^- + IO_3^-; I_2 by ID	8P
	RC	Monograph	1L
Antimony			
^{124}Sb	M, RM	Copptn. with MnO_2	36B
	M	Study of electrodepos.	22D
	RM	Distribution in steel	13G
	M	Solv. extn.	7R

*RR = radioreagent; ID = isotope dilution; RM = radiometric; M = method study; RC = radiochemical.

†These numbers are references in the original article, which see.

A fruitful application of radiometric analytical techniques is in problems where speed and precision are necessary for adequate quality control. A recent study explores such problems in the cement industry and offers a family of methods using radioisotopes to solve some of them. These techniques and others are discussed in the following paragraphs, which also summarize recent papers covering the determination of fluoride by an extraction procedure using ^{182}Ta, of sulfate with ^{51}Cr, and of several metals by amalgam-exchange separation.

Table I-2 TYPICAL ORGANIC APPLICATIONS OF RADIOTRACERS

Material determined	Tracer	Method*	Remarks	References†
Acids	^{14}C	ID	In irradiated $\phi COOH$	19D
"Aerosol"	^{35}S	RM	Conc. at liq. surfaces	26D
Alcohols	^{14}C	ID	Chrom. on SiO_2	8E
Aldosterone	^{3}H, ^{14}C	RR, ID	Acetyl. with ^{3}H-Ac_2O	53K
Alkaloids	^{32}P	RR	Paper chrom., "devel." by ^{32}P-molybdophos.	73S
Amino acids	^{14}C	ID	Chrom. sepn.	10W
Amino acids	^{3}H, ^{14}C	ID, RR	Acetyl. with Ac_2O	8W
Amino acids	^{131}I	RR	With ^{131}I-pipsyl chloride	28K, 29K
Alanine, γ-aminobutyric acid	^{131}I, ^{35}S	RR, ID	Pipsyl chloride and ^{35}S cpd.	29K, 4U
Benzylpenicillin	^{14}C	ID	^{14}C cpd., hyd. to ϕCH_2COOH	38A

*RR = radioreagent; ID = isotope dilution; RM = radiometric.

†These numbers are references in the original article, which see.

Radiometric Analytical Methods in Quality Control

Cement manufacturers require rapid feedback of analytical information to maintain a product within specifications. Since the raw materials are quite variable, the process depends heavily on chemical analysis; but some of the analytical methods, although fairly well standardized, are lengthy or laborious.

Workers at Battelle Memorial Institute (BMI),[3–5] under a Division of Isotopes Development (DID) contract, have developed radiometric methods for determination of several of the more important constituents of cement, i.e., magnesium, calcium, sulfur, iron, and aluminum. These methods appear to be the equal of standard American Society for Testing and Materials (ASTM) techniques in rapidity or, in the case of lengthy methods such as those for magnesium, calcium, or sulfate, considerably better. The experimental techniques are relatively simple and could be applied to similar industries having the same analytical problems in quality control, such as in the manufacture of glass or lime. The only specialized equipment required is a scaler and a Geiger-Mueller (G-M) dip tube.

Analysis of cement by neutron activation, also attempted by these workers, was only semiquantitative, although such a method is quite suitable for determination of trace elements.

The methods developed for magnesium, sulfur, and calcium were radiometric adaptations of the conventional gravimetric methods, in which a compound of the desired element is precipitated. Iron and aluminum, on the other hand, were found to lend themselves to a modified complexometric titration, with radiotracers replacing the usual colorimetric or potentiometric end-point indicators.

MAGNESIUM

Method. The radiometric method for determining magnesium oxide in cement has been submitted to the ASTM for adoption.[3] Basically,

it is the same method as that outlined in the ASTM standards on cement,[6] the only modification being the labeling of $(NH_4)_2HPO_4$ with ^{32}P. The labeling makes possible a direct determination of the amount of precipitate formed without carrying out the usual steps of digestion, filtration, ignition, and weighing. A similar radiometric titration was developed by Langer[7] about 20 years ago, with Na_2HPO_4 as a titrant. However, that procedure requires the addition of precipitant to an ammoniacal magnesium solution, and, as pointed out by Willard and Furman,[8] the addition of phosphate ion to such a solution tends to promote the incorporation

Table I-3 SCHEME FOR RADIOMETRIC ANALYSIS

Ions sought	Radioactive reagent	Comments
Al, Zn,* Mg, Mn,* Ca	$(NH_4)_2H^{32}PO_4$	*p*H must be adjusted within proper limits; usually reduced or elevated temperatures required; digestion period uncertain; coprecipitation of foreign ions should not interfere; buffering of solutions should not interfere unless buffering agent reacts with radioactive reagent
Al, Ga, In, Ce, La, Zr, Bi, Fe(III), U, Th, Be	$Na_2H^{32}PO_4$	
CrO_4^{2-},* Mo, VO_4^{3-}, AsO_4^{3-}, Cl^-,* Br^-, I^-,* CN^-,* CNS^-	$^{110}AgNO_3$	*p*H must be adjusted in some cases; reactions proceed at room temperature and are fairly rapid; lower limit of sensitivity of ions that form complexes or weak acids is higher than others
Sc, V, rare earths, Pb, Ca, Au	$H_2{}^{14}C_2O_4$	
K, Tl	$Na_3{}^{60}Co(NO_2)_6$	
F^-,* PO_4^{3-}, CO_3^{2-}, $C_2O_4^{2-}$	$^{45}CaCl_2$	*p*H must be adjusted within proper limits
BrO_3^-; IO_3^-, ClO_3^-, ClO^-	$K^{131}I$	Phase separation dependent on extraction into CCl_4 of $^{131}I_2$ formed
Sb, As, Fe, Mn, Cr, V, Mo, K, $C_2O_4^{2-}$, other organic compounds	^{144}Ce-^{144}Pr	Extraction of Ce(IV) into dioctyl phosphoric acid in *n*-heptane; growth of daughter must be allowed for
Al, Be, P, B, Mn, Ni, U(VI), Fe, V, Zr, Tl, Sn, Ta, Ga	Cupferron-^{14}C	
Fe, V, Zr, Tl, Sn, Ta, Ga, Cu, Cd, V, Al, Zn	8-hydroxyquinoline–^{14}C	Separates Mg from alkali and metals and Al from certain elements and complex ions, e.g., Mg, Be, PO_4^{3-}, AsO_4^{3-}, B, F, Mo, Ti, W, Ta; adjustment to proper *p*H necessary for efficient precipitation

*Driscoll et al.[2] performed these analyses. They also determined the following ions (radioreagents in parentheses and brackets): Pb^{2+}, $Ba^{2+}(Na_2{}^{35}SO_4)$; $Cl^-[^{203}Hg(NO_3)_2]$; $SO_4^{2-}[BaCl_2(^{35}SO_4)]$; and Ni^{2+} (dimethylglyoxime with ^{63}Ni as indicator).

of foreign ions in the precipitate, causing high results.

In the BMI work the apparatus shown in Fig. I-1 was used for determining MgO by precipitation as $MgNH_4PO_4$. Other equipment needed is a 100-ml syringe, which is used to draw the solution into the chamber, and a counting instrument to register the radioactive disintegrations picked up by the G-M tube.

In testing the method, solutions of $(NH_4)_2HPO_4$ and MgO were prepared at concentrations of 4 and 1 mg/ml, respectively. The $(NH_4)_2HPO_4$ solution was labeled with sufficient $^{32}PO_4^{3-}$ to give initial counting rates of 7000 to 8000 counts/min, and 10 ml of this reagent was added to a solution containing about 10 mg of MgO. The mixture was diluted to 100 ml, 3 drops of methyl red indicator were added, and the whole was cooled in an ice bath to below 4°C with continuous stirring. The frit of the apparatus was placed so that it would still be below the solution level when the sample chamber was full. The sample chamber and tube were flushed with the solution and then refilled, and the radioactivity level of the solution was measured. The precipitation was brought about by drop-by-drop addition of concentrated ammonia, with stirring, to the methyl red color change; 10 ml of excess ammonia was then added. After 5 min stirring at a temperature below 4°C, the sample chamber and tube were again flushed with the solution, the chamber was refilled, and the radioactivity of the solution was again measured. The decrease in the radioactivity level was a direct measure of the amount of phosphate reacted to form $MgNH_4PO_4$. The measurement was corrected for background, and the radioactivity after precipitation was corrected for dilution by the added ammonia. A sample calculation follows:

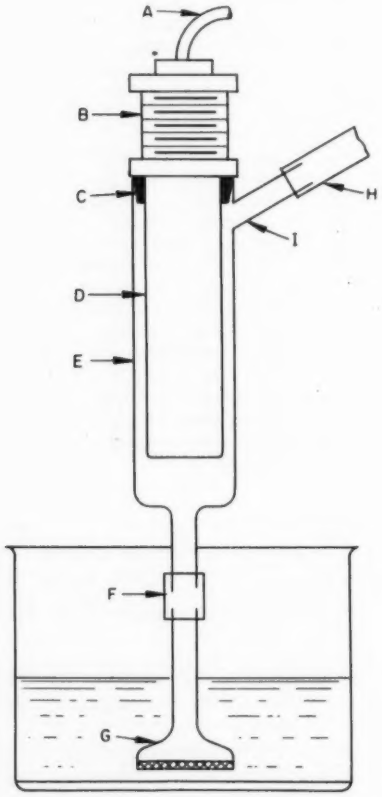

Fig. I-1 Radiometric titration apparatus.

A High-voltage lead
B G-M tube connector and socket
C Shaped rubber stopper
D G-M tube, thin-wall aluminum type 1B85, Victoreen Instrument Co.
E Glass solution chamber
F Rubber tubing
G Coarse glass frit
H Rubber tubing to 100-ml syringe
I Glass chamber sidearm

Initial net radioactivity	7280 counts/min
Final net radioactivity	2925 counts/min
Initial $(NH_4)_2HPO_4$	40.0 mg
Final $(NH_4)_2HPO_4 = 40 \times 2925/7280$	16.2 mg
$(NH_4)_2HPO_4$ reacted	23.8 mg
Mol. wt. MgO/mol. wt. $(NH_4)_2HPO_4$	40.3/132
Weight of MgO = $23.8 \times 40.3/132$	7.27 mg

Results. The results of 16 determinations at various MgO concentrations between 5 and 25 mg per 100 ml of solution showed a standard deviation of <0.14 mg of MgO and a relative error of <0.7%. For smaller samples the relative error was larger (Table I-4). Experiments to determine the effect of several interfering elements, such as aluminum, calcium, iron, manganese, sodium, and potassium, showed that calcium and manganese were the only ones affecting results. Manganese is present in such low concentrations in cement that it presents no problem. Calcium, however, as a major constituent of cement, must be removed before the radiometric determination of magnesium. (In the current ASTM procedure, also, this separation must be made during the complete analysis of cement.) Since a principal advantage

Table I-4 PRECISION AND ACCURACY OF MgO DETERMINATIONS AS A FUNCTION OF MgO CONCENTRATION

Magnesium (as MgO), mg				
Added	Found	Average	Standard deviation	Relative error, %
0.97	0.90			
	0.95			
	0.89	0.91	0.033	−6.19
4.85	4.86			
	4.88			
	4.84			
	4.82	4.85	0.026	0.00
9.70	9.93*			
	9.68			
	9.70			
	9.72			
	9.78	9.72	0.043	+0.21
24.25	24.51			
	24.55			
	24.32			
	24.31	24.42	0.138	+0.70

*Determination excluded since its deviation from the average of the other four is more than four times their average deviation.

of the radiometric method is speed, the magnesium should first be rapidly and quantitatively separated from the other cations in cement. An ion-exchange method was developed by these workers which permits complete separation of magnesium from aluminum, calcium, and iron (see Ref. 5 for details).

CALCIUM

Method. In tests on determination of calcium, $Ca_3(^{32}PO_4)_2$ was precipitated from an alkaline solution by $(NH_4)_2HPO_4$ labeled with ^{32}P. The method and calculations were similar to those used for the magnesium.

Results. The results of 21 calcium determinations, ranging from 3 to 33 mg per 50 ml of solution, showed a relative error of <1% for CaO concentrations of 13 to 33 mg per 50 ml—again with larger errors for smaller concentrations (Table I-5). These determinations were made with a constant initial ratio of $(NH_4)_2HPO_4$ to CaO of 2.4/1. The following elements were used in interference studies: manganese, potassium, aluminum, magnesium, and iron. The last three were found to interfere with the determination of calcium, and the first two did not interfere.

Attempts to determine calcium by complexometric titration were unsuccessful.

SULFUR

Method. Sulfur was determined by precipitation of sulfate with $Sr(NO_3)_2$ tagged with ^{89}Sr. Acetone was added to the solution to decrease the solubility of $SrSO_4$ in the precipitation system. In tests on the method, mixtures of K_2SO_4 and $Sr(NO_3)_2$ were stirred and digested on a steam bath for 10 min. The mixture was allowed to cool, portions were centrifuged, and 5-ml aliquots of the supernatant liquid were counted. The decrease in the counting rate of the supernatant liquid as compared to the original labeled $Sr(NO_3)_2$ was taken as a measure of the strontium removed. The counting rates were corrected for background and dilution, and results were calculated similarly to those for magnesium.

Results. The results of sulfate analyses were comparable to those for magnesium and calcium (Table I-6) for a series of determinations in which 30.8 mg of K_2SO_4 per sample (14.2 mg of SO_3) was present.

Table I-5 PRECISION AND ACCURACY OF CaO DETERMINATIONS AS A FUNCTION OF CaO CONCENTRATION

Calcium (as CaO), mg				
Added	Found	Average	Standard deviation	Relative error, %
3.33	2.94			
	2.83	2.88	0.020	−13.5
8.33	7.86			
	7.98	7.92	0.027	−4.9
10.00	9.31			
	9.91			
	9.89	9.70	0.34	−3.0
13.32	13.36			
	13.30			
	13.17	13.28	0.031	−0.3
16.66	16.63			
	16.60			
	16.81			
	16.80			
	16.63			
	16.60			
	16.67			
	16.63	16.67	0.027	+0.06
33.32	33.26			
	33.92			
	33.39	33.52	0.35	+0.6

Table I-6 PRECISION AND ACCURACY OF SULFATE DETERMINATIONS

Sulfate (as K_2SO_4), mg				
Added	Found	Average	Standard deviation	Relative error, %
30.8	30.7			
	31.0			
	30.8			
	30.8			
	30.1			
	30.7			
	30.7			
	30.0*			
	31.0	30.7	0.028	−0.33
18.5	18.2			
	18.9			
	18.6	18.6	0.35	+0.54
3.7	4.1			
	3.9			
	3.3	3.8	0.41	+2.6

*Not included in average since the experimental conditions were different from those of the other determinations.

IRON

Method. A technique was developed for determining iron by complexometric titration with ethylenediaminetetraacetic acid (EDTA), in which $^{91}Y_2(C_2O_4)_3$ was used as the radiometric end-point indicator. In the tests, 25 ml of Fe(III) solution (20 to 40 mg of Fe_2O_3) and 7 to 14 ml of 1% tartaric acid solution (70 to 140 mg) were mixed and buffered at *p*H 5 with 10 ml of 1*M* sodium acetate solution. About 200 mg of solid $Y_2(C_2O_4)_3$ labeled with ^{91}Y was added, and the mixture was stirred 10 min and then counted. Counting rates, corrected for dilution and background radiation, were plotted against the volume of EDTA added. The end point of the titration was determined by back-extrapolating the line drawn through the experimental points beyond the end point until it intersected a horizontal line determined from the original counting rate (Fig. I-2).

Results. The results of tests on the radiometric titration of 15 and 37 mg of Fe_2O_3 with 0.05*M* EDTA at *p*H 5.0 indicated a standard deviation of <0.3 mg of Fe_2O_3 and relative errors of about 1.5 and 0.3%, respectively (Table I-7), in initial volumes of 50 ml. The determinations were all made with constant mole ratios of Fe_2O_3 to tartaric acid and to sodium acetate of about 3.6 and 43, respectively.

ALUMINUM

In preliminary work on a back-titration method for determining aluminum, a solution containing aluminum was buffered at *p*H 9.5 with a NaOH–boric acid mixture. A known excess of EDTA and an excess of $Na_3{}^{32}PO_4$ were added to the mixture. The excess EDTA was back-titrated with standard $BaCl_2$ solution, and the formation of insoluble $Ba_3(^{32}PO_4)_2$ was used to indicate the end point. The results of these experiments indicated that the accuracy would not be high, and work was suspended.

Complexometric titrations with radiometric end points were also studied for consecutive determination of iron and aluminum in the same solution, but the results showed that aluminum interfered with the determination of the iron end point. The procedure involved titration of the iron in solution at *p*H 5 with standard EDTA, $^{91}Y_2(C_2O_4)_3$ being used as the radioactive end-point indicator. This was followed by titration of the aluminum at *p*H 9 with standard EDTA, with $^{110}AgIO_3$ as the end-point indicator.

Radiotracer Method for Determining Fluoride

The extensive use of fluoride chemicals today — in fluoridation of water, in rocket fuels, and in pollution control of waste streams and

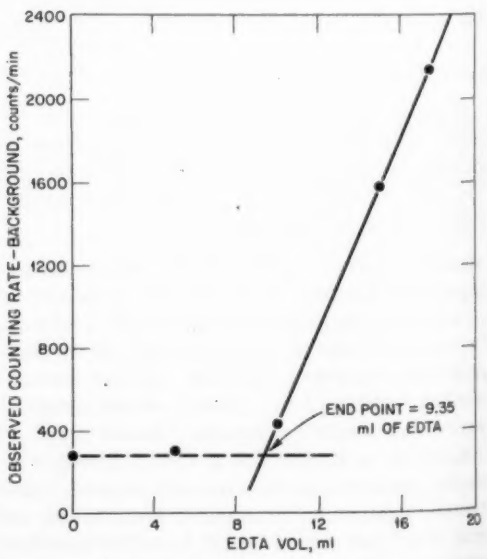

Fig. I-2 Radiometric titration of iron.

air—has stepped up the quest for fast, accurate analytical methods. Most current methods are either time-consuming or poorly selective, or they involve "negative measurements"; that is, the fluoride ion bleaches a metal-dye indicator, for example, and the decrease in color is measured,[9] whereas previous methods using radiotracers[10,11] depended on measuring the decrease in radioactivity after a reaction. Belcher, Leonard, and West[12] have developed a positive colorimetric method using hydroxyanthraquinones.

Moore,[13] working at ORNL under a DID contract, recently reported a relatively sensitive and simple positive radioreagent method. In Moore's technique, ^{182}Ta (half-life, 115 days) is extracted as a fluoride complex from an aqueous system, under highly selective conditions, into an organic solvent. The amount of radioactivity extracted is proportional to the fluoride-ion concentration. Several appealing features of this approach over current methods are speed, high selectivity, adaptability to colored solutions, high tolerance for oxidants, and absence of need for critical pH or temperature control.

METHOD

The solution used in Moore's work had the composition:

^{182}Ta: 4.5×10^5 gamma counts/(min)(ml)

H_2SO_4: 17.8M

Total Ta: <1.0 μg/ml

The procedure consists in pipetting an aliquot of fluoride solution into a 1-oz polyethylene bottle and then adding the appropriate amounts of distilled water, 18M sulfuric acid, and 1 ml of the ^{182}Ta radioreagent to produce a final 5-ml 6.5M aqueous solution of sulfuric acid. After the solution is mixed by gentle swirling, it is extracted for 2 min with 5 ml of diisobutyl ketone reagent. The contents are transferred to a 15-ml glass centrifuge tube and centrifuged for 2 min in a clinical centrifuge. One milliliter of the organic (top) phase is carefully pipetted

Table I-7 PRECISION AND ACCURACY OF Fe_2O_3 DETERMINATIONS

Fe_2O_3 added, mg	EDTA conc., M	Mole ratio: Fe_2O_3 to tartaric acid	Mole ratio: Fe_2O_3 to sodium acetate	Fe_2O_3 found, mg	Experimental average, mg	Standard deviation, mg of Fe_2O_3	Relative error, %
7.36	0.0200	3.60	43.2	7.50			
				7.61	7.56	0.08	+2.72
14.72	0.0200	3.60	43.2	14.72			
				15.09			
				15.04	14.95	0.20	+1.56
	0.0499	3.60	43.2	14.52			
				14.68	14.60	0.11	−0.82
36.81	0.0499	3.60	64.8	37.02			+0.57
			64.8				
			21.6				
			43.2				
			21.6				
37.12	0.0499	3.60	43.2	36.82			
				37.14			
				37.29			
				37.22			
				37.33			
				37.81			
				37.17			
				37.02	37.22	0.29	+0.27
73.62	0.0499	3.60	43.4	74.13			
				76.82	75.48	1.9	+2.52
		2.88	35.8	67.26			−8.65
		1.94	21.6	*			

*End point could not be observed.

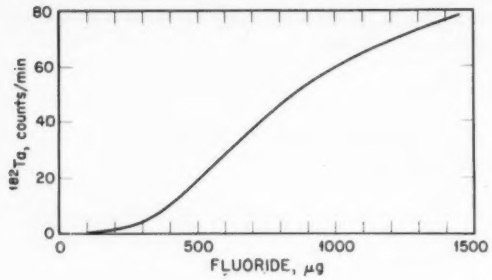

Fig. I-3 Standard calibration curve. Extraction of ^{182}Ta from 6.5*M* H_2SO_4 as a function of fluoride concentration is shown.

into a 10- by 75-mm culture tube, the tube is stoppered, and the gamma activity is counted in a well type scintillation counter. The fluoride-ion concentration is determined by correcting the counting rate for background, blank, and decay, and then referring to a standard calibration curve (Fig. I-3).

The calibration curve is prepared by running 400-, 500-, 600-, 700-, 800-, and 900-μg portions of standard fluoride solution through the above procedure. If it is desired to use the lower, nonlinear, part of the curve, 100-, 200-, and 300-μg portions of standard fluoride are run similarly. A new calibration curve should be run when fresh radioreagent is prepared.

The radioreagent, ^{182}Ta, is readily available in high purity from ORNL and without special license in 10-μc amounts.

Details of preparation of reagents are given by Moore.

DEVELOPMENT OF THE METHOD

Radioreagent Solutions. Antimony-125 and ^{95}Nb were considered as radioreagents in addition to ^{182}Ta. The first tracer was essentially inextractable by various solvents from a sulfuric acid–hydrofluoric acid medium. Niobium-95 tracer is extracted efficiently from sulfuric acid–hydrofluoric acid solutions, but a relatively high concentration of hydrofluoric acid must be present.

A concentrated sulfuric acid system was selected as the best because high extractability of ^{182}Ta with concomitant high selectivity is possible. In addition, sulfuric acid was found to markedly enhance the extraction of tantalum-fluoro species (Table I-8). Tantalum-185–sulfuric acid solutions were found to be stable for at least 5 months.

Extractants. Diisobutyl ketone and methylisobutyl ketone were excellent extractants for tantalum-fluoro species from sulfuric acid solution. Diisobutyl ketone was selected because of its greater immiscibility and inertness at higher sulfuric acid concentrations and the low blanks possible. Negligible sulfuric acid was found to be extracted into diisobutyl ketone; however, the solvent was pretreated in all cases by mixing well for 2 min with an equal-volume portion of 0.5*M* H_2SO_4.

An interesting observation was that one may use an organic-to-aqueous ratio as low as 1 to 10 and still achieve the recovery possible with a 1-to-1 ratio. This fact is potentially useful for concentration purposes with very low levels of fluoride.

Experiments showed the desired inextractability of ^{182}Ta tracer with diisobutyl ketone from aqueous solutions of varying sulfuric acid concentration up to 7*M*. Less than 0.2% extraction was found, thereby ensuring low blanks in this system. It is recommended that a blank determination (no fluoride) be run when fresh reagents are prepared. Blanks usually averaged 0.04%, or 40 gamma counts/min per milliliter of extractant.

Table I-8 EFFECT OF SULFURIC ACID CONCENTRATION ON EXTRACTION OF ^{182}Ta TRACER FROM DILUTE HF SOLUTION

H_2SO_4, *M*	HF, *M*	^{182}Ta extracted, %
5.3	0.0049	8.34
6.5	0.0008	0.20
	0.0049	19.15
7.5	0.0008	0.6
	0.0049	31.2

Mechanism. The mechanism is of the ion-association type and probably proceeds by the following oxonium type reactions:

$$^{182}Ta_2(SO_4)_5 + 14HF \rightleftharpoons 2H_2{}^{182}TaF_7 + 5H_2SO_4 \quad (1)$$

$$H_2{}^{182}TaF_{7(a)} + 2R_2CO_{(o)} \rightleftharpoons [(R_2COH^+)_2][{}^{182}TaF_7^{2-}]_{(o)} \quad (2)$$

where the subscripts *a* and *o* refer to aqueous and organic phases, respectively. Equilibrium is reached in 2 min or less. Extraction of radiotantalum with nonoxygenated solvents, such as xylene or chloroform, is negligible.

Effect of Diverse Ions. Several of the major interferences of current procedures were tested at the 700-μg fluoride level. Table I-9 shows the limiting amounts of several of these ions. Each value is the average of two or more determinations. Sulfate ion, a common interference in most fluoride methods, is a requirement in this new method. The method shows marked tolerance to anions, in general, as a result of the high acidity (6.5 *M* sulfuric acid) from which the extraction is performed. Thus repression of the ionization of weak acids, such as acetic, citric, tartaric, and oxalic, renders them relatively innocuous. The orthoborate ion was the major anionic interference tested. Among the cations tested, tolerance was highest for thorium, with relatively good tolerance to the usual strong fluoride-complexing ions, zirconium, iron, and aluminum. A positive bias was noted for the higher levels of iron and calcium tested. Calcium precipitated at the 1-mg level. The uranyl ion interfered seriously, showing a positive bias even at the 60-μg level.

Table I-9 INFLUENCE OF VARIOUS ANIONS AND CATIONS ON FLUORIDE RECOVERY

Ion	Amount added	Recovery, %
Tartaric acid	0.255 millimole	97
Citric acid	0.264 millimole	98
Oxalic acid	0.168 millimole	99
Acetic acid	1.00 millimole	100
Phosphoric acid	0.17 millimole	96
	0.27 millimole	90
	0.34 millimole	90
Boric acid	0.009 millimole	56
Nitric acid	0.50 millimole	103
	1.00 millimole	118
Hydrochloric acid	0.50 millimole	91
	1.00 millimole	88
Th(IV)	0.56 mg	102
	1.12 mg	103
	1.68 mg	102
Zr(IV)	0.10 mg	100
	0.21 mg	95
	0.31 mg	90
	1.03 mg	62
Fe(III)	0.10 mg	103
	0.30 mg	108
	0.50 mg	122
	1.00 mg	152
Al(III)	0.11 mg	90
	0.34 mg	78
	0.57 mg	66
Ca(II)	0.20 mg	109
	0.50 mg	153
	1.00 mg (precipitates)	150
UO_2(II)	0.012 mg	92
	0.030 mg	101
	0.060 mg	127

Table I-10 DETERMINATION OF FLUORIDE IN SOME TYPICAL SAMPLES

Sample*	Fluoride found, μg: By thoron method	Fluoride found, μg: By radiotracer method
NaF	300	290
	500	488
	700	700
HF	980	1005
Process sample 1	1020	1034
Process sample 2	640	590
Process sample 3	780	822

*Process samples were dark-yellow and contained approximately 150 μg of niobium per milliliter.

Results. In Table I-10 some typical determinations of fluoride ion by this procedure are compared to those by the standard thoron method.[14] In these tests the counting rate was linear over the range 400 to 840 μg of fluoride ion; below 400 μg it was nonlinear but reproducible and useful. The sensitivity was about 40 μg of fluoride ion.

Determination of Sulfate with ^{51}Cr

A straightforward analytical procedure to determine sulfate with ^{51}Cr as a radiotracer, recently developed by Armento and Larson at ORNL,[15] is applicable to a wide range of concentrations of sulfate. At higher concentrations it is comparable to gravimetric and volumetric methods for sulfate; the practical lower limit is about 10^{-3} millimole in a 25-ml volume, equivalent to 4 μg/ml. This method can be used to determine sulfate in the presence of substances that interfere with a normal redox titration. The combination of radiotracer and computer calculation described in this paper is applicable to specialized work in which analysis is required of large numbers of relatively noncomplex samples of sulfate of varying concentrations. This technique uses the well-known reaction in which the sulfate is precipitated with excess HCl solution of $BaCr_2O_7$. After neutralization of the solution with am-

monia, the insoluble $BaSO_4$ and excess $BaCrO_4$ are filtered off, and the soluble chromate, equivalent to the sulfate originally present, is determined by redox titration with, for example, Fe(II). Instead of being titrated, however, in this modified procedure the excess chromate is determined by measuring its activity, $BaCr_2O_7$ labeled with ^{51}Cr (half-life, 27.8 days, gamma emitter) having been used as the precipitant.

Sixty-one samples were counted in five series of tests (8 to 14 in each series), the sulfate concentration being varied from 0.25×10^{-4} millimole to 2.5 millimoles. A well type scintillation counter with a 2- by 2-in. NaI(Tl) crystal was used to count gamma radiation (10^4 counts per sample). A least-squares analysis of each series of data was made on an IBM-7090 computer. Agreement between the observed counts and the value calculated from the least-squares analysis was excellent. The relative error was well within 3% for most of the data. Reliable results were obtained at sulfate concentrations so dilute that the reaction could not be followed visually. The method is theoretically usable to the limit set by the solubility of $BaSO_4$, but many factors pertaining to its insolubility affect reproducibility. Preliminary work suggests that addition of organic solvents such as alcohols, which lower the solubility of $BaSO_4$, markedly decreases the error of the method at extremely low concentrations.

For unknown sulfate concentrations the procedure involves analysis of aliquots of several standard sulfate solutions so as to bracket the unknown concentration, if possible. The activity (in counts per minute) is plotted against concentration. Approximately 10^4 counts are accumulated, and background is determined to ensure absence of extraneous activity. The unknown value is calculated by using the activity-concentration curve obtained above.

The only interferences with this method are anions which form with barium precipitates that are about as soluble as $BaCrO_4$, e.g., $BaNH_4PO_4$.

Amalgam-Exchange Technique for Separating Radioactive Elements

When several radioactive elements occur in the same solution, it is often necessary to separate them and to determine the amounts of the separated elements by counting their activities. An amalgam-exchange technique has been developed by Meinke and coworkers at the University of Michigan for separating radioisotopes of Cd,[16] In,[17] Sr,[18] Bi,[19] and Tl (Ref. 20) from contaminating radioactive elements. Most of the separations take less than 10 min to perform. In their procedure there are two isotopic-exchange steps:

Table I-11 SEPARATION OF METALS BY AMALGAM EXCHANGE*

Element	Yield of desired element, %	Time, min
Bi	97.5	10
Tl	95	6
In	95	11
Cd	80	8
Sr	34	8

*Yield of contaminants, in most cases, was less than 0.1%.

1. An aqueous solution of the desired radioactive element, *M, is contacted with the nonradioactive form of the same element in amalgam form;

$$M(Hg) + {}^*M^+ \rightleftharpoons {}^*M(Hg) + M^+$$

The necessary condition for the isotopic exchange is that the number of atoms of the element in the amalgam phase be much greater than that in the aqueous phase.

2. The element is back-extracted into an aqueous phase, and the activity is then counted by ordinary methods.

The procedure is simple and, although the kinetics are not always clear, the conditions are altered so that maximum separation of the desired element is obtained in each step.

In the Michigan work, various metals were studied. The degree of separation and yield of each element obtainable were measured with radioactive tracers, and many factors that affect the procedure, e.g., concentrations of reagents, interferences, and time of agitation, were studied and evaluated. The studies on bismuth and thallium, the procedures for which are among the best examples of selective separation by this technique (Table I-11), are described here as representative of separations by the amalgam method.

Separations in each instance were made in 0.5-oz Boston round bottles with polyethylene insert screw caps. The bottle was clamped to a mechanical shaker to which a 12-in.-long extension arm was connected for added radial action. Gross-gamma-ray measurements were made with a Nuclear-Chicago proportional counter as described elsewhere.[21] For thallium, gross-beta-ray measurements were made with the same counter, and beta-ray spectrum measurements were made with a hollow Scintilon plastic phosphor coupled with a RIDL 400-channel analyzer; gamma-ray spectrum measurements were made with a special 400-channel gamma-ray scintillation spectrometer.

BISMUTH

Materials:

Mercury (Baker and Adamson analytical reagent, triple-distilled, further purified by washing with dilute nitric acid and rinsing with distilled water)

Nitrogen gas, > 98%

Bismuth metal sticks, c.p.

Saturated copper sulfate solution in 1*M* nitric acid, Baker analyzed reagent

All other nonradioactive reagents were of c.p. or analyzed reagent grade; the radioactive tracers listed in Table I-12 have been described previously[21]

Preparation of Amalgam. The amalgam is made by adding the appropriate amount of metal (1.4 wt.% bismuth dissolves in mercury at room temperature) to the mercury stored under 0.5*M* HNO_3 in a beaker. The system is heated and agitated to amalgamate the bismuth completely.

Procedure. To 2 ml of a well-mixed 0.25*M* H_2SO_4 solution containing 10 μg of bismuth metal and interfering ions is added 200 μl (~2.7 g by weight) of 1.4% bismuth amalgam (~38 mg of bismuth). The system is purged with nitrogen gas for 1 min, and the bottle is quickly capped and shaken for 2 min. The aqueous phase is removed, and the amalgam is transferred to another bottle, washed twice with 2-ml portions of 0.1*M* HNO_3, and then transferred to a third bottle containing 2 ml of

Table I-12 SEPARATION OF BISMUTH, THALLIUM, AND INTERFERING ELEMENTS BY AMALGAM EXCHANGE*

Tracer	Weight,† μg	Reduction potential,‡ volts	Element separated,§ %					
			Thallium determination			Bismuth determination		
			Exchange	Back-exchange	Total	Exchange	Back-exchange	Total
^{137}Cs	C.F.	−2.92	0.01	N.D.¶	<0.01	<0.01	N.D.	<0.01
^{140}Ba-^{140}La	5.2	−2.90, −2.52	0.0082	51.8	<0.005	<0.003	N.D.	<0.002
^{90}Sr-^{90}Y	C.F.	−2.89, −2.37	0.012	N.D.	0.01	0.005	N.D.	<0.002
^{144}Ce-^{144}Pr	C.F.	−2.48, −2.47	0.011	N.D.	<0.01	<0.01	N.D.	<0.01
^{95}Zr-^{95}Nb	C.F.	−1.53, −1.1	0.0025	N.D.	0.002	<0.01	N.D.	<0.002
^{65}Zn	180	−0.76	0.004	N.D.	0.002	0.002	N.D.	<0.002
^{131}I	C.F.	**	0.012	N.D.	0.011	35.8	0.17	0.06
^{131}I	10^3 of ^{127}I	**				0.09	N.D.	<0.003
$^{115}Cd^m$	24.8	−0.40	10	0.1	0.01	15.5	0.075	0.01
$^{114}In^m$	4.9	−0.34	0.01	7.5	<0.01	0.006	N.D.	0.005
^{204}Tl	20	−0.34	97		95.5	0.002	N.D.	<0.002
^{60}Co	14	−0.28	0.028	41	0.01	<0.003	N.D.	0.002
^{113}Sn	33.5	−0.14	0.20	20.5	0.04	<0.004	N.D.	0.001
^{124}Sb	1.3	+0.21	0.045	17.2	<0.01	0.05	10.3	0.005
^{210}Bi	510	+0.32	55.5	0.02	0.011	97.5	99	97
^{106}Ru-^{106}Rh	C.F.	+0.60, +0.25	13	0.42	0.05	14.7	0.7	0.01
^{75}Se	8	+0.74	27.2	5.3	1.45	27	0.55	0.15
^{192}Ir	50	+0.77	1.84	0.8	0.013	1.4	0.7	0.01
^{203}Hg	3.8	+0.79	40.1	0.02	<0.01	34.3	<0.03	<0.01
$^{110}Ag^m$	40	+0.80	99.6	0.1	0.1	96	<0.1	<0.1

*Average of duplicate runs except for bismuth and thallium, each of which is the average of five runs.

†Weight of inactive element present before separation. C.F. = carrier-free.

‡Standard reduction potential of lowest stable oxidation state to the elemental state. Data taken from Latimer.[22]

§Elements are listed in order of their reduction potentials.

¶Not detectable since no measurable radioactivity was in amalgam.

**Iodine in its lowest reduced state.

saturated cupric sulfate solution in 1*M* HNO_3. This bottle is capped and shaken for 5 min. A 100-μl aliquot of the aqueous layer containing the separated bismuth is removed and counted. Inactive bismuth was used for interference studies, and radioactive bismuth and nonradioactive interfering substances were used for yield determinations.

THALLIUM

Materials:

Mercury (Baker and Adamson analytical reagent grade, further purified by shaking with 2*M* nitric acid, followed by rinsing with distilled water)

Nitrogen gas, water pumped, 99.9% pure

Thallium metal sticks, c.p.

Thallous acetate, c.p.

All other nonradioactive reagents were of c.p. or reagent grade; the radioactive tracers listed in Table I-12 have been described previously[21]

Preparation of Amalgam. The amalgam is made by washing thallium metal with 2*M* HNO_3 until the surface is completely etched. It is then washed with distilled water and dried with tissue papers. An appropriate weight of thallium metal (underwater, purged with nitrogen) is added to 10 g of mercury, and the whole is stirred to amalgamate the metal completely.

Procedure. To 2 ml of a well-mixed 0.5*M* $NaNO_3$ solution containing 10 μg of thallium metal and interfering ions is added 100 μl (1.3 g) of thallium amalgam containing 3 wt.% thallium (~40 mg). The system is purged with N_2 for 30 sec; then the bottle is quickly capped and agitated for 1 min. The aqueous layer is removed, and the amalgam is transferred to another bottle, washed twice with 2-ml portions of distilled water, and then transferred to a new bottle containing 2 ml of thallous acetate solution. After a 4-min agitation, 100 μl of the aqueous phase containing separated thallium isotopes is evaporated on a planchet and then counted.

Table I-13 TEMPERATURE DEPENDENCE OF BISMUTH SEPARATION ON TEMPERATURE AND AGITATION TIME

Temp., °C	Agitation time in extraction, min	Exchange, %	Agitation time in back-extraction, min	Total yield, %
52	0.5	84.3	0.5	70
	1	95.8	1	87.3
	1.25	97.5	2	88
90	0.25	91.1	0.25	69.2
	0.5	98.0	1	90.6
			1.5	94.8
			2	96.7

Table I-14 DEPENDENCE OF THALLIUM YIELD ON AGITATION TIME IN NITROGEN-PURGED AND -UNPURGED SYSTEMS

Agitation time, min	Exchange, %	
	Unpurged system	Purged system
0.5	63	96
1.0	85.8	97
1.5	86	96.5
2.0	85.5	94
3.0	59	92
4.0	49	89

RESULTS AND DISCUSSION

In preliminary experiments performed to determine the optimum separation procedure, yields for bismuth improved with increasing temperature and longer agitation time (Table I-13). For thallium, shorter agitation times could be used at higher temperatures to produce equilibrium—only 2.5 min at 50°C compared to 4 min at room temperature for both metals.

The yields were dependent upon purging the system with nitrogen. For example, the results for thallium are shown in Table I-14. Yields also depended on agitation time in both the exchange back-extraction steps (Table I-12). With the exception of cadmium and iodine, elements more electropositive than bismuth (see Table I-12) are not carried over in the initial exchange step of bismuth separation. Only cadmium among the elements above thallium is carried over in this step for thallium. Iodine is decreased appreciably when 1 mg of iodide carrier is added to the exchange medium for bismuth separation. With both bismuth and thallium, cadmium is probably reduced and amalgamates with mercury. The elements below bismuth and thallium do contaminate the amalgam-exchange step owing to the reduction of their ions by the amalgam. With bismuth the activities remain in the amalgam and are not extracted into the cupric solution, which therefore selectively removes bismuth from the

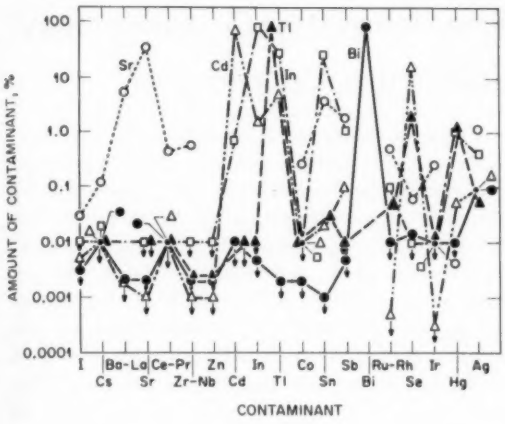

Fig. I-4 Decontamination of strontium, cadmium, indium, thallium, and bismuth in radiochemical separations by amalgam exchange. Arrow indicates separation better than measurable lower limit.

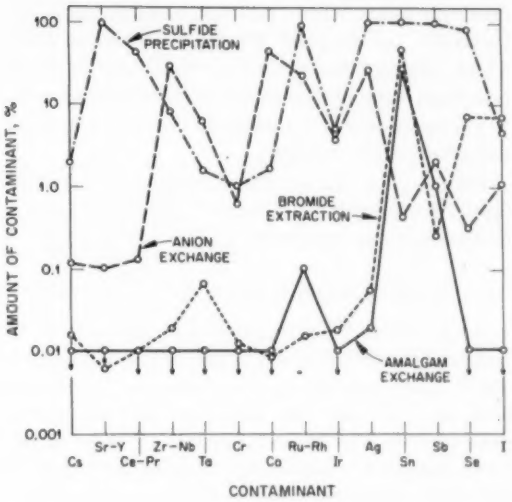

Fig. I-5 Contamination of indium separated by four different methods. Recovery by sulfide precipitation, 92%; bromide extraction, 93%; anion exchange, 96%; amalgam exchange, 97%. Arrow indicates separation better than measurable lower limit.

Table I-15 DETERMINATION OF RADIOSTRONTIUM IN RAINWATER SAMPLES

Sample No.	Counting time, min	Counts	Net counts/min	Amount of radiostrontium,* μc/ml	Average of duplicate runs, μc/ml
1	30†	528	16.3	1.09×10^{-7}	
					1.07×10^{-7}
		510	15.7	1.05×10^{-7}	
2	35†	539	14.1	0.94×10^{-7}	
					0.93×10^{-7}
		529	13.8	0.92×10^{-7}	
3	50‡	525	9.3	0.60×10^{-7}	
					0.62×10^{-7}
		546	9.7	0.64×10^{-7}	

*Standard deviation is about 5%.
†Background = 1.3 counts/min.
‡Background = 1.2 counts/min.

Table I-16 DETERMINATION OF GROSS ACTIVITY IN RAINWATER SAMPLES*

Sample No.	Counting time, min	Counts	Counts/min			Volume of sample, ml	Gross activity, μc/ml
			Total	Background	Net		
1	50	565	11.3	1.2	10.1	20	5.7×10^{-7}
2	50	510	10.2	1.2	9.0	20	5.1×10^{-7}
3	80	488	6.1	1.2	4.9	20	2.75×10^{-7}

*Standard deviation of the counting rate is about 5%.

mercury phase. For thallium, considerable selectivity is obtained by using thallous ions, which preferentially oxidize thallium from the amalgam without oxidizing the elements below it.

Yields also varied with volume of amalgam and of exchange medium.

Figure I-4 shows the decontamination obtained for thallium, bismuth, and other metals in separate experiments.

COMPARISON WITH OTHER SEPARATIONS

Ruch et al.[17] compared the separation of indium by amalgam exchange with other methods.[23] The results, shown in Fig. I-5, indicate that, in general, the amalgam method gives somewhat better decontamination than bromide extraction and considerably better decontamination than anion exchange or sulfide precipitation. It is more convenient since it avoids the necessity of working with flammable ethers and since the higher immiscibility results in phase separation.

APPLICATIONS

The amalgam separation method is generally useful in analyzing samples for the metals mentioned; in addition, some separations have specific applications. For example:

1. The amalgam-exchange method is simpler and faster for the separation of thallium from fission products than the thallium chromate method.

2. In the determination of thallium by neutron activation analysis, short-lived ^{206}Tl (half-life, 4.2 min) can be used. The amalgam-exchange procedure gives a high decontamination for thallium and requires only ~5 min at 50°C.[24]

3. The amalgam-exchange method has been used to analyze samples of rainwater for radiostrontium content by an indirect method involving the precipitation of strontium on calcium carbonate carrier. Several samples were analyzed by Qureshi and Meinke.[18] All samples were counted through a Mylar window of 0.9 mg/cm^2 thickness on a Tracerlab low-level-beta counting system. Table I-15 summarizes the analysis of 500-ml rainwater samples, and Table I-16, the determination of gross activities by evaporation of a total of 20 ml in 2-ml portions on a stainless-steel planchet. Analysis of other environmental samples, such as milk or grass, required long preparation time and was not considered practical with this method.

New Edition of Scintillation Spectrometry Gamma-Ray Spectrum Catalogue

A new edition of the *Scintillation Spectrometry Gamma-Ray Spectrum Catalogue*, which has been in preparation for the last two years, is being issued.[25] This edition is a complete revision of the original data compilation,[26] which was issued in 1957. Like the original catalog, the new edition contains a collection of spectra representing the response of a standardized scintillation spectrometer to the gamma radiation emitted by individual radioactive nuclides. Figure I-6 is an example of a typical curve. In addition to the graphs representing the response of a 3- by 3-in. NaI detector in a standard geometrical arrangement, the data are also presented in digital form for the preparation of punched-card or paper- or magnetic-tape libraries for data analysis (Fig. I-7).

The 1964 catalog is in two loose-leaf volumes and contains data for over 250 isotopes. All spectra have been normalized to a standard set of gain scales, and a comprehensive text is presented which describes the fundamentals of gamma-ray scintillation spectrometry; the details of spectrometer design, electronic systems, and instrumental calibration; and data reduction.[27–33] An extensive bibliography is included which lists current literature in all phases of this field.

An important addition to the catalog is the extension to include spectra of neutron-deficient isotopes. This phase of the work has been carried out with the support of the AEC Division of Isotopes Development. This addition to the original data collection was considered most important in view of the increasing application of accelerator-produced isotopes.

An extensive index is included to facilitate the use of these data, with separate tables of data listed according to gamma-ray energy (major gamma rays), half-life, method of production, and specialized categories. In its present form the new edition of the catalog represents the initial collection of data for a continuing data service. As new or improved data become available on different isotopes, this information will be made available in the form of supplements to the original issue.

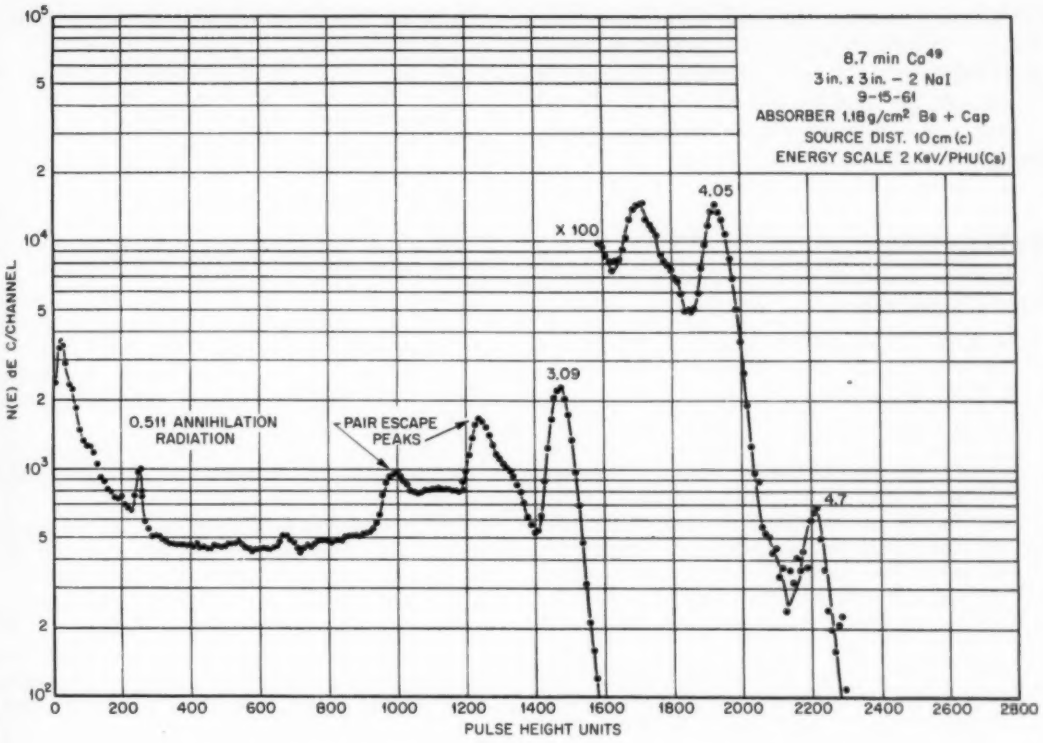

Fig. I-6 Sample of a typical spectrum curve from *Scintillation Spectrometry Gamma-Ray Spectrum Catalogue*. The original spectrum size was 11 by 17 in.

8.7 MIN. CALCIUM 49

DETECTOR 3X3-2 DATE 9-15-61

PLATE NO. 20-49-1 SOURCE DISTANCE 10 CM.

ENERGY SCALE 2.0 KEV/PHU ABSORBER 1.18 G/CM SQ. BE + CAP

INTENSITY NORMALIZATION NONE

	0	1	2	3	4	5	6	7	8	9
000	000000	023770	033959	034630	028566	023557	022103	018321	014933	013166
010	012615	012579	011840	010593	009142	008792	008146	007818	007447	007394
020	007440	006966	006721	006600	007500	009700	009900	007900	005416	005088
030	005040	005044	004946	004790	004736	004719	004623	004629	004643	004600
040	004700	004600	005386	004700	004500	004482	004466	004641	004594	004579
050	004569	004662	004694	004671	004795	004695	004495	004481	004359	004450
060	004400	004460	004410	004440	004500	004620	004800	005106	005119	004867
070	004732	004493	004490	004296	004544	004602	004521	004627	004821	004868
080	004804	004748	004825	004887	004826	005008	005031	005025	005064	005052
090	005197	005211	005346	005453	005782	006292	007637	008717	009362	009514
100	009705	009346	008865	008486	008071	007947	007858	007973	008097	008083
110	008222	008197	008248	008099	008086	008021	008052	007981	008206	008850
120	009790	011512	013714	015525	016511	016101	015244	014079	012711	011694
130	011081	010599	010242	009803	009405	008578	007993	007143	006193	005697
140	005243	005364	006338	008845	012409	016802	020405	022322	022656	020355
150	017235	013339	009715	006932	004731	003121	002162	001609	001211	000971
160	000942	000859	000805	000741	000817	000829	000922	001046	001235	001365
170	001403	001422	001440	001248	001173	001120	001052	000876	000821	000787
180	000748	000690	000661	000583	000497	000498	000490	000508	000590	000764
190	000977	001175	001344	001429	001335	001235	001075	000835	000681	000501
200	000363	000264	000191	000128	000096	000088	000056	000052	000051	000043
210	000045	000034	000037	000029	000036	000032	000041	000036	000044	000037
220	000050	000055	000058	000040	000036	000024	000020	000016	000021	000023
230	000011	000006	000003	000009	000007	000012	000008	000007	000004	000000

Fig. I-7 Sample of a typical page of digital data from *Scintillation Spectrometry Gamma-Ray Spectrum Catalogue*.

The second edition has been issued as USAEC Report IDO-16880 and will receive standard distribution in the *Physics* category. Copies will be offered for sale by the Office of Technical Services, U. S. Department of Commerce, Washington, D. C., 20230.

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Section

II

Isotopes and Radiation Technology

Radiation Processing of Foods

In earlier issues of *Isotopes and Radiation Technology*, applications of gamma rays to the processing of foods have been discussed. The following two articles discuss more fully some of the work sponsored by the U. S. Atomic Energy Commission in this area. The first, dealing with the Marine Products Development Irradiator, is a facility study detailing the design of the building, the source, and the device for continuous irradiation of certain seafood products. The second, covering an investigation of the effect of gamma radiation on the palatability and general acceptability of a number of raw and blanched seafoods, was carried out at the Bureau of Commercial Fisheries at Gloucester, Mass., with irradiations done at the Massachusetts Institute of Technology irradiator.

Marine Products Development Irradiator

By Philip Miller and R. J. Herbert*

A recent survey[1] describes the widespread interest and effort in the use of high-energy ionizing radiation as a processing tool. One of the chief applications is in the preservation of food.[2,3] The purpose of this paper is to describe a specific project in this area, the Marine Products Development Irradiator (MPDI), which utilizes the gamma rays (1.33 and 1.17 Mev) of ^{60}Co.

The two important approaches to preserving food against bacterial action by radiation are through sterilization and pasteurization. The former is comparable to the conventional canning process, and the latter, to the heat pasteurization of milk. Table II-1 shows the chief characteristics of these two methods. Other processes, generally using low doses (10,000 to 50,000 rads) and not involving antibacterial action, include inhibition of sprouting of potatoes and onions, control of trichinosis in pork, deinfestation of grain and flour, and control of ripening in bananas and other fruits.[4]

Since 1960 the U. S. Army has been emphasizing sterilization, and the AEC, pasteurization. The Army work is now being concentrated at its Radiation Laboratory in Natick, Mass.[5] In February 1963 radiation-sterilized canned bacon was approved by the Food and Drug Administration for human consumption, and testing is under way on chicken, ham, and other food items. The AEC efforts were mentioned earlier.[3c]

Food preserved by irradiation, like that preserved by heating or freezing, may show changes in flavor, color, texture, and nutritive value. These changes vary considerably with different foods, and, as might be expected, they are affected more by large than by small amounts of radiation; thus certain foods are logical candidates for sterilization and others for pasteurization.

Marine products are being considered for radiation pasteurization for several reasons:

1. The refrigerated shelf life of seafood can be extended from a week to a month or more, without significant effects on flavor or nutritive value, by doses of 100,000 to 500,000 rads. This has been shown in research at the Massachusetts Institute of Technology,[6] the University of Washington,[7] and the Bureau of Commercial Fisheries in Gloucester. At Louisiana State University, research is being conducted on shrimp. However, sterilization doses may have serious effects on flavor.[8]

2. Tripling the shelf life should make possible shipment of refrigerated fish economically to any part of the country and thus greatly expand the market. The consumption of fresh fish is now largely confined to coastal areas.

*Associated Nucleonics, Inc., Garden City, N. Y.

Table II-1 COMPARISON OF STERILIZATION AND PASTEURIZATION

Characteristic	Sterilization	Pasteurization
Dose, Mrads	2.5 to 5	0.25 to 0.50
Bacteria population	Eliminated	Decreased
Containers	Hermetically sealed	Not hermetically sealed
Storage	Room temperature	32–35°F
Shelf life	Indefinite	3- to 5-fold increase

3. The pasteurization process can be introduced with little disturbance to the normal commercial procedures for distributing fish. Thus, after the fish have been filleted and packed, they can be routed through an irradiation cell before being distributed.

Description of the MPDI

The MPDI is being built as part of the AEC's program for developing and demonstrating the technology of radiation processing in food preservation. It is located in Gloucester, Mass., at the site of the Technological Laboratory of the Bureau of Commercial Fisheries, U. S. Department of Interior. Construction started in July 1963, and the plant is expected to start operation about August 1964. It will be operated by the Bureau, under an agreement with the Division of Isotopes Development of the AEC, to investigate the pasteurization of seafoods on a semicommercial basis. The MPDI will be used to process finned fish, such as haddock, and shellfish, such as clams. The primary difference in handling these seafoods is in the type of container used, the fillets being in rectangular tins holding a maximum of 30 lb and clams in No. 10 cans. The contract specified that the basic design should emphasize handling the fillet tins.

A photograph of a scale model of the one-story MPDI building is shown in Fig. II-1, and Fig. II-2 is a top view. At the left is a general office and working area of about 2500 sq ft; the smaller area at the right is the gamma cell with a gross area, including the shield walls, of 1250 sq ft. The general area includes the lobby, office, laboratory for health physics and dosimetry, cold storage room, filleting area, and conveyor loading area.

Figure II-3 is a sketch of the storage pool, elevator, and source plaque. The storage pool is a 16-ft stainless-steel tank containing 15 ft of water, in which the source is stored in a vertical position. The elevator controlling the source operates somewhat like an overhead garage door and is placed in its operating position (horizontally, between the upper and lower tracks of the conveyor) inside an aluminum shroud. In this position it is cooled by a stream of air blowing through the shroud.

Description of the Source

DESIGN

Preliminary studies were made to determine the absorption characteristics of the fillet packages for various geometric source-package arrangements. The studies were based on three sizes of commercial fillet tins, all 11 by 15 in., ranging in thickness from $2\frac{3}{4}$ to $6\frac{1}{8}$ in., and holding 10, 20, or 30 lb of fish. Two 20-lb tins handled as a 40-lb unit were also considered.

Ideally, the fish should receive the same dose at the surface as at the center for homogeneous pasteurization. But the surface always receives more than the center owing to absorption, and the ratio of the maximum to minimum doses is always greater than 1. Thus the dose at the center necessary for pasteurization controls the total dose required. Because off-flavor at the surface becomes a problem at dose ratios greater than about 1.3, that figure was chosen as the highest allowable ratio. It was found that a single 30-lb tin was the thickest unit with which this maximum-to-minimum dose ratio requirement could be met.

By use of a target overlap approach—surrounding the source as much as possible with target material for most efficient use of the source energy—it was determined that four passes across the source were the maximum number that still fell within the 90-min time limitation set on the process (see below, "Design Changes for Commercial Unit"). The maximum-to-minimum dose ratio at the package center line was estimated to be 1.29; the required source strength, 205,600 curies; and the utilization efficiency, as defined below, 20.7%. The product within the tin was assumed to be homogeneous in making the calculations, and the dose rate in the packages in any plane parallel to the source plane was assumed to be unaffected by the gaps and excess metal between the packages.

The utilization efficiency, E_u, is defined as the ratio of energy absorbed in the product at the

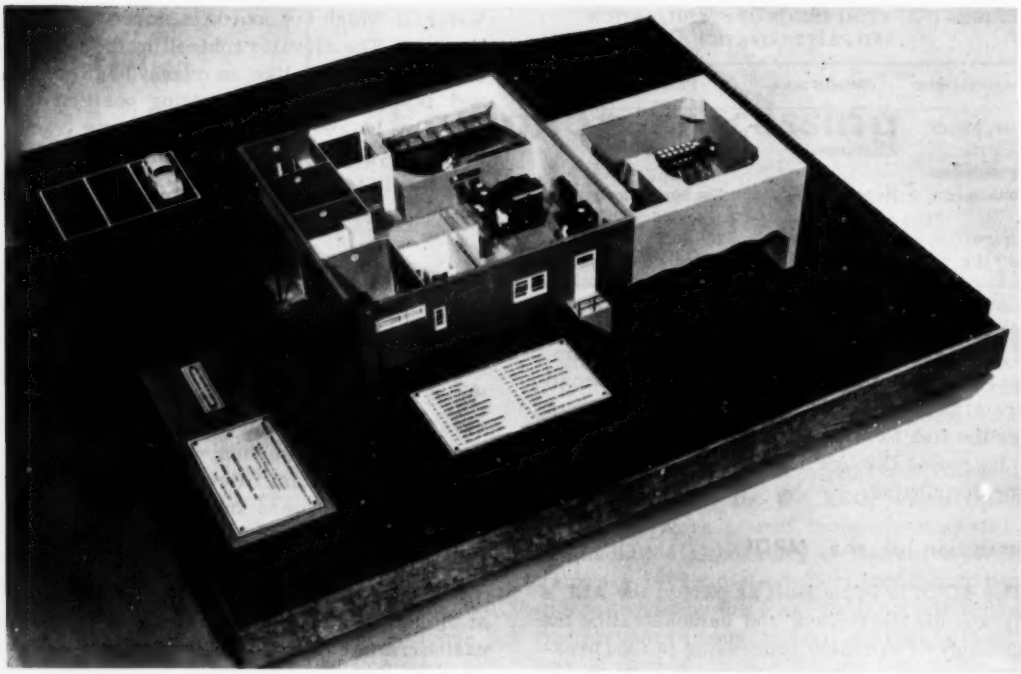

Fig. II-1 Scale model of MPDI building (side view).

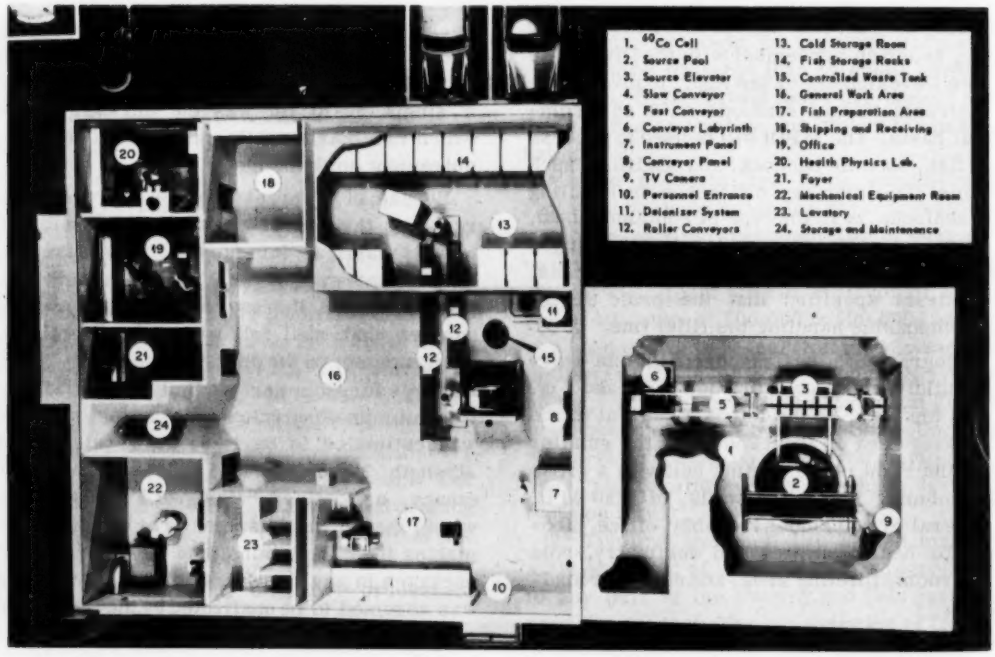

Fig. II-2 Scale model of MPDI building (top view).

minimum specified dose to the total electromagnetic energy emitted from the source. For ^{60}Co,

$$E_u = \frac{85 \times \text{Mrad-lb/hr}}{\text{No. of curies in source}}$$

The integrated dose to a target point traversing the source was assumed equal to the dose received at that point if it were stationary in front of the plane source having a length at least equal to the length of travel.[9] For plane sources of this size, Manowitz et al.[10] showed that increasing the plane length beyond 50 in. had very little effect on the centerline dose up to 6 in. away from the source plane. Therefore, introducing and removing the package at least 2 ft away from the closest edge of the source would effectively maintain dose uniformity in the product.

Absorption coefficients for the product were assumed to be those of water with suitable density corrections. To account for parasitic absorption in the source, shroud, and intervening structural material, a radiation-absorber thickness equivalent to 0.3 of a relaxation length (i.e., 0.3 of the thickness of absorber required to reduce by a factor, e, the dose from a hypothetical point source) was assumed in estimating the dose-rate values in the product. This assumption is conservative in estimating the source strength but provides flexibility in adjusting the maximum-to-minimum dose rate by the addition of absorbers between the source and the target.

The calculations showed that the near-optimum source width (normal to the package movement) is the width of the package itself. The model can then be represented by a package being irradiated by a source twice its width.[11] Since the package width is 11 in., the apparent source is 22 in. wide. Manowitz et al.[10] have presented centerline data for a target having the same absorption characteristics as water with a 4-in. source overlap, a close approximation to the 5-in. overlap of the proposed 22-in. source. To evaluate the dose characteristics in the product, Manowitz's data were adjusted to apply to a fillet density of 0.83 g/cm^3. Figure II-4 presents the

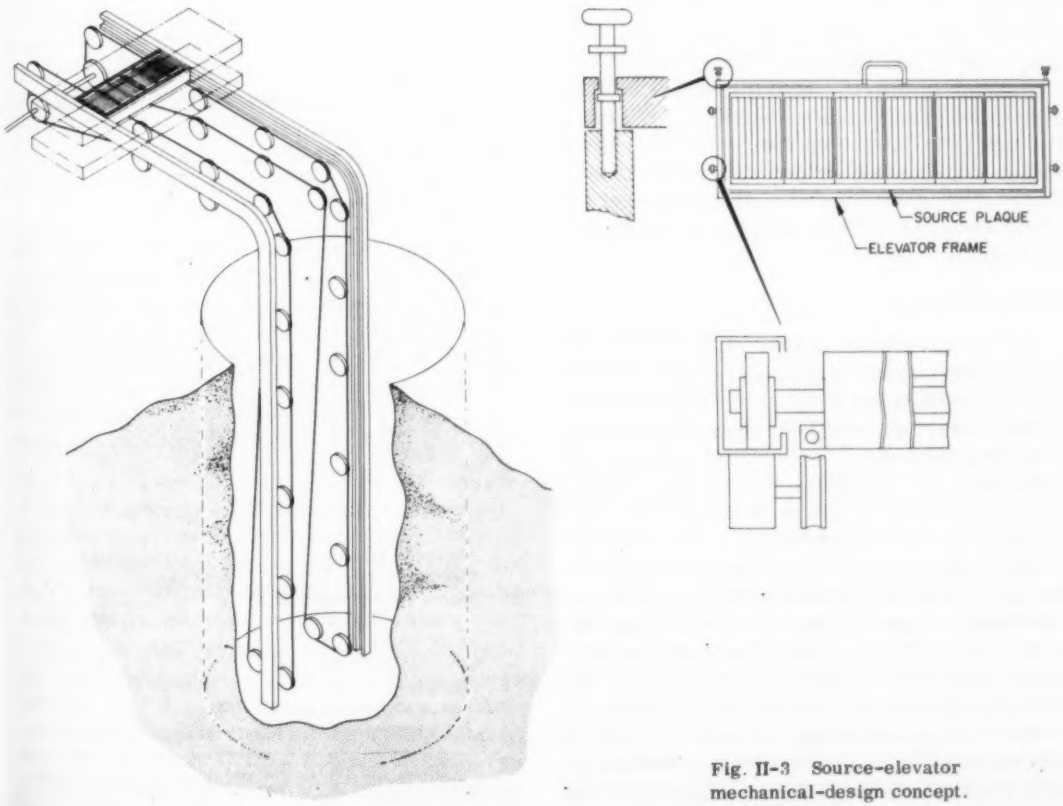

Fig. II-3 Source-elevator mechanical-design concept.

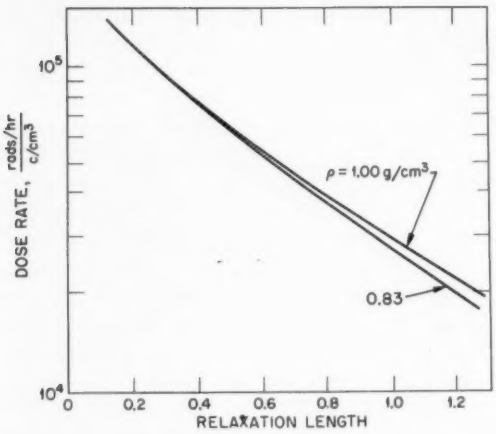

Fig. 4 Depth-dose variation in water type target from ^{60}Co slab source.[10]

adjusted curve. A detailed description of the mathematical procedures used is presented elsewhere.[9]

Inherent in the calculations is the assumption that the source is homogeneous. However, with package motion normal to the strip length, each source strip acts essentially independently; thus variations in specific activities from strip to strip (50% variation is common) will not affect the flux pattern. The specific activity along the length of the source strip is expected to be constant; if it is not, any variation may be effectively averaged by alternating the orientation of each strip.

CONSTRUCTION

The source plaque is approximately 1 by 4 ft and is made up of six subunits. These subunits were assembled in a hot cell at Brookhaven National Laboratory (BNL) and shipped in two 7-ton lead shipping casks[12] to Gloucester. They were loaded into the plaque in the storage pool with long-handled tools. Table II-2 lists the chief characteristics of the source.

The cobalt strips were contained in welded stainless steel and irradiated at Savannah River; they were enclosed at BNL in a second stainless-steel jacket. Thus the outer jacket is nonradioactive, and, even if some corrosion occurs in the pool, no radioactivity will get into the water.

The product normally receives a dose of 500,000 rads at a production rate of 1000 lb/hr with a maximum-to-minimum ratio of 1.3. The dose can be decreased to 150,000 rads or less if desired, first by increasing the conveyor speed and then by removing one or more of the six sections into which the source is divided.

The cell has ordinary concrete walls more than 5 ft thick and a 4-ft-thick roof with a removable plug for lowering the cobalt shipping cask into the pool. The cell operation is viewed remotely by television. There are a number of safety interlocks to prevent accidental exposure of personnel to radiation, and strategically placed openings in the walls and roof can be used to introduce long-handled tools should the elevator malfunction. As another safety measure the ^{60}Co cell is kept at a lower pressure than the rest of the building. Air is drawn from the building into the cell through the labyrinth and is exhausted through an absolute filter to a stack.

Shielding Design

The *Code of Federal Regulations*[13] was used as a basis for providing adequate personnel protection against radiation. Shielding study was required in three areas: (1) water pool storage, (2) wall and ceiling thicknesses, and (3) labyrinth designs. The assumptions made were: a point source having a strength of 300,000 curies of ^{60}Co (although this leads to a slightly optimistic appraisal of the collided flux, the saving in computation time derived from this assumption is warranted since the effect is small); no parasitic absorption by material in the cell.

The minimum water-level requirement, calculated to be 15 ft, was determined by the once-a-year procedure of loading and unloading a shipping cask at the bottom of the pool. The source is shipped in units containing one-sixth of the total source (or a maximum of 50,000 curies). After removal from the cask, the top of a unit is about 11 ft from the pool surface, leav-

Table II-2 CHARACTERISTICS OF ^{60}Co SOURCE

Amount of activity	250,000 curies*
Specific activity	50 curies/g
Source strip dimensions, doubly encapsulated	13.5 by 0.8 by 0.15 in.
No. of strips	84
Self-absorption for double-layer source	15%
Overall source utilization efficiency	21%

*Allows for 16 months' decay before falling below design rating.

ing about 1 ft of water in excess of that needed to meet the protection criteria. The ^{60}Co photons of energy 1.17 and 1.33 Mev were considered individually in making the computations, and the attenuation coefficients and dose buildup factors presented by Grotenhuis[14] were used.

The following factors were considered in estimating the required ceiling and wall thicknesses:

1. Unlimited access criteria[13] would prevail in the general work area opposite the interior (east) wall and at ground level adjacent to the three exterior cell walls (Fig. II-2).
2. The roof would be a restricted area where the maximum dose level would be about 100 mr/hr. Radiation at ground level due to "skyshine" was estimated to be negligible.

The attenuation of ^{60}Co gamma rays in concrete was calculated,[15] and the required thickness of the east wall was found to be 5 ft 6 in. The outside walls could be made 5 in. thinner, since the site contours are such that personnel can approach the walls only from well below the level of the irradiation chamber. The roof requires only a 4-ft thickness.

There are two entrances to the ^{60}Co cell, one for personnel and the other for product transfer. To provide easy access, the personnel entrance combines a partial labyrinth with a manually operated shielding door. The product conveyor, by necessity, passes through an open labyrinth. The dose levels in each labyrinth were estimated by the albedo concept.[9,16] The gamma-ray deflection pattern in the labyrinths was simulated by considering exposed areas condensed to a point for the emitted portion of the gamma energy. To use this method, an accurate scale drawing was employed for each labyrinth design studied.

Many gamma-ray paths are possible, but only those paths were considered which permitted the least number of pseudoreflections. Since the albedo coefficients for concrete as presented by Perkins[17] range from 10^{-2} to 10^{-3} for most events, the result of neglecting the more than minimal deflected gamma-ray paths would amount to less than 10% of the dose.

The dose rate calculated at the entrance to the product labyrinth by the albedo concept was less than 0.1 mr/hr, ignoring the conveyor and other equipment installed in the labyrinth. Unpublished data in the authors' files obtained at the large ^{60}Co cell labyrinth in the irradiation facility at Natick, Mass., indicated that this approach may be optimistic by a factor of 4. This is explained in part by some observations by Terrell et al.,[18] which illustrate that albedos for concrete increase with decreasing energy since energy degradation was not considered in the calculations. Even after this factor has been applied to the calculations, the dose level is still acceptable under the federal guidelines.

Several open and closed labyrinths that would not increase the size of the cell were considered for personnel entrance. The closed labyrinth was chosen because the open one required both additional space and elaborate safety devices.

A dose level of 2 rads/hr at the entrance of the personnel labyrinth was effectively reduced to a tolerable value by the addition of an 18-in.-thick concrete door. Gamma rays reaching the door will already have been scattered through more than 100°. According to the National Bureau of Standards Handbook[15] values for 90° scattered rays, the thickness calculated was 18 in. of concrete of 1.6 in. of lead. From an evaluation of the energy dependence on scattering angle presented by Allen,[19] 15 in. of concrete was calculated. A concrete thickness of 14 in. was estimated by the curves presented by Mooney and Braestrup.[20]

Labyrinth calculations at best give only an approximation to the dose rates anticipated in any design. Few data exist for albedo coefficients, and most of the available data are for thin slabs. In view of the dearth of present-day knowledge, a conservative design approach was taken.

Processing Procedure

In the normal operation of the MPDI, fillet tins are brought to the building by truck and are placed in the cold storage room (32 to 35°F) on movable racks. However, as an alternative procedure, the filleting can be done inside the building, which contains filleting tables and a waste-disposal unit. The cold storage room can hold a total of 12 tons of fish. The packages are transferred on a gravity roller conveyor from the cold storage room to the feed point for the irradiator. A high-speed mechanical conveyor, moving at 50 ft/min, carries them into the gamma cell through a vertical labyrinth that goes under the floor. Figure II-5, a diagram of the conveyor arrangement, shows a round trip of two packages. Inside the cell the packages are transferred to a

slow-speed conveyor, moving at 0.75 ft/min, which carries them past the radiation source. Since the packages have loose-fitting covers and must be kept in a horizontal position, the source is also horizontal.

Both conveyors operate on a start-stop basis to permit loading and unloading and transfer between the fast and slow conveyors. Two packages are carried side by side on a single carrier, or basket, which is part of the conveyor. Each package makes a round trip under and over the source on one side of the source center line and is only half irradiated after one trip. It then comes out of the cell, is shifted to the other side of the carrier by the operator, and then goes back into the cell for a second round trip. In each round trip the first pass is under and the second is over the source so that, in case of any dripping, the falling liquid will be more completely irradiated than the package it falls on. This ensures that none of the completely irradiated packages can become contaminated with partially pasteurized liquid.

The carriers are almost touching each other, as mounted on the conveyor, so as to minimize loss of radiation. Thus the packages follow each other closely in the direction of travel. The slow conveyor is loaded with packages for a distance of 9 ft, compared with a source length of 4 ft. In the other direction a double row of packages is 22 in. wide compared with a source width of 11 in. (based on the bare cobalt). Thus packages extend beyond the source in both directions to improve the efficiency (as discussed more fully above under "Description of the Source").

After a package has been completely irradiated, which takes about 1 hr, it is returned to the cold storage room. Each package bears a tab that changes color from yellow to red after two round trips in the irradiator. The number of passes is indicated by paint markings automatically applied to the tin.

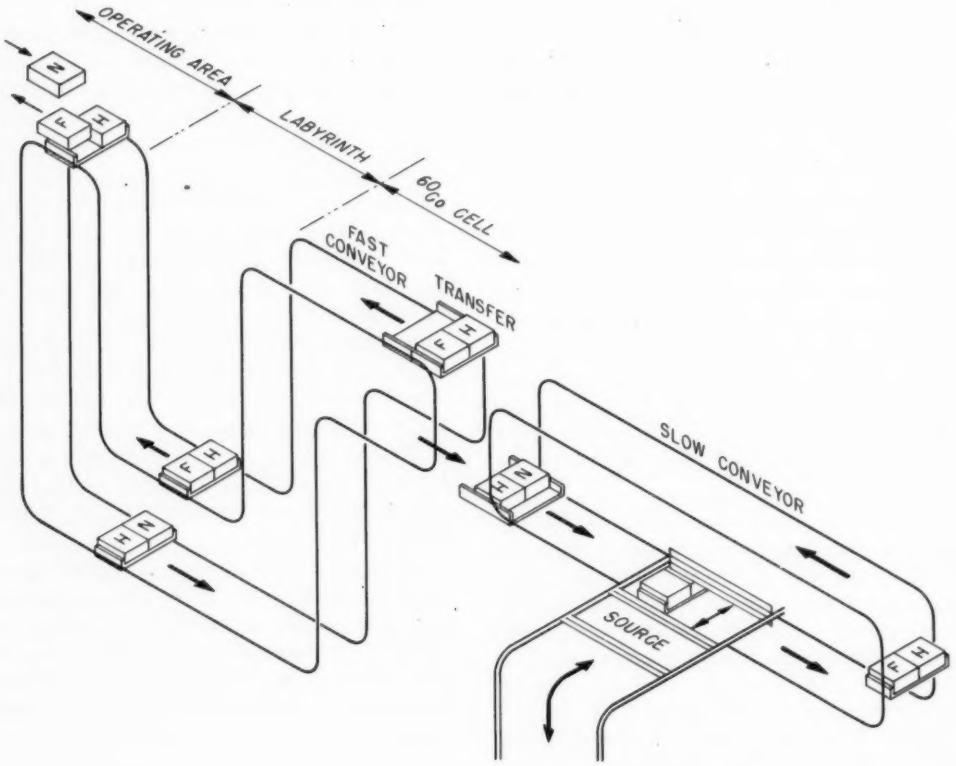

Fig. II-5 Schematic drawing of conveyor system.

Design Changes for Commercial Unit

The capacity and essential characteristics of the MPDI are close enough to those of a full-sized plant to give a convincing demonstration of the commercial feasibility of fish pasteurization. At the same time, because of budget limitations and because it is intended primarily as a development unit, at least two of its features are probably unsuited for a commercial plant:

1. It was arbitrarily decided that the product need not be refrigerated during irradiation and that the maximum time it could be kept out of the cold storage room is 90 min. A separate high-speed conveyor was provided to cut down the residence time in the labyrinth to a few minutes, and the source utilization efficiency is limited to about 20% (as described above) since there is only enough time for the packages to make four trips past the source. A commercial production unit would be designed to keep the product under refrigeration at all times, and the number of passes would then be increased to at least eight, possibly more, giving a source utilization efficiency of 30 to 40%. The decision on whether to use a high-speed conveyor in the labyrinth would be based only on equipment cost and inventory, not on residence time.

2. The size of the cell is larger than is needed for a commercial unit. The MPDI is designed to have some degree of flexibility so that it can be used for batch experiments with large packages and for other special tests. The use of wet storage for the source is also partly to give more flexibility in handling. In a commercial unit the smaller cell would become essentially a concrete shield closely surrounding the conveyor and package array. Dry storage for the source could be considered. Commercial operation would be continuous, and the conveyor might have an automatic feed-storage unit that would require little labor on the two night shifts.

Economics

Based on the approaches just described, a preliminary estimate of the cost of pasteurizing fish in a commercial plant has been made. Assuming a production rate of 2000 lb/hr, a minimum dose of 500,000 rads, and an average utilization efficiency of 50%, the cost is estimated at 2.6 cents per pound. There are recent indications[3] that a dose of 250,000 rads may be adequate, in which case the estimated cost becomes 2.0 cents per pound. The Bureau of Commercial Fisheries has made some surveys[21] which indicate that pasteurization costs in the range 1 to 5 cents per pound might be commercially attractive.

Radiation Preservation of New England Seafoods

By Joseph W. Slavin, Maynard A. Steinberg, and Louis J. Ronsivalli*

Microbiological changes proceed rapidly in chilled seafoods and contribute greatly to loss of quality and onset of spoilage.[22] Haddock, ocean perch, pollock, clams, and many other fish and shellfish caught off the New England coast have a limited storage life in ice, 6 to 12 days, the exact time varying with the particular species and the conditions of storage.[23] Because of the lack of a suitable method of extending the quality of fresh seafood, markets for fish in this form are limited to cities close to the major fishing ports. Freezing and canning permit distribution of large quantities of fish and shellfish to inland areas, but these processed products differ in appearance and taste from those in the fresh state. Thus a satisfactory means of extending the refrigerated shelf life of fresh fish without adversely affecting quality will be of significance to both producers and consumers.

In 1959 Carver and Steinberg[24] investigated the application of gamma rays for this purpose, with screening studies to determine the acceptability of raw and blanched cod, pollock, whiting, butterfish, and flounder irradiated at levels ranging from 116,500 to 1,860,000 rads. Their work showed that the refrigerated storage life of butterfish and cod was significantly increased without adverse quality changes by irradiation of 300,000 to 400,000 rads. Some species were more amenable to this process than others, and low dose levels and refrigerated storage resulted in a significant extension of the shelf life of some fishery products. They also observed that in many cases doses of over 500,000 rads resulted in off-odors and -flavors in the irradiated product.

In 1960 Proctor, Goldblith, and Nickerson made an evaluation of the technical, economic, and practical feasibility of radiation preserva-

*Bureau of Commercial Fisheries Technological Laboratory, Gloucester, Mass.

tion of fish for the AEC.[25] They reviewed past work on the irradiation of seafoods and concluded that doses of less than 1,000,000 rads would successfully prolong the shelf life of many species of fish and shellfish when stored at temperatures of 32 to 40°F. They also presented a comprehensive program of research and development for introducing radiation-pasteurized fish into the U. S. economy. Subsequently, the AEC's Division of Isotopes Development initiated a research program on the preservation of seafoods with gamma rays and refrigeration.

Also in 1960 studies were undertaken at the Bureau of Commercial Fisheries Technological Laboratory in Gloucester, Mass., on irradiation of marine fish and shellfish of the northwest Atlantic. The specific objectives of this research were to: (1) study the effects of irradiation and storage on nutritional value, as judged by changes in amino acids and B vitamins; (2) investigate the flavor and odor changes that take place in the irradiated product; (3) determine radiation dose levels suitable for extending the refrigerated shelf life of soft-shell clam meats and fillets of haddock, pollock, and ocean perch; and (4) measure the effect of preirradiation quality on post-irradiation storage life of the product. Irradiations were carried out at the MIT ^{60}Co facility, a BNL Mark I food irradiator with a source energy of approximately 30,000 curies.

This report reviews some of the more significant findings[26] of these investigations.

Effect of Irradiation on Nutritional Value

The effect of low levels of radiation on nutritional content is illustrated by our study on soft-shell clams. Comparison of the free and total amino acids and B vitamin contents of irradiated samples with those in commercially processed samples showed no great differences. Irradiation alone at a level of 4,500,000 rads resulted in a 40% increase in the free amino acid content of the raw clams (Table II-3), but the free amino acid contents of the raw fresh clams varied greatly, from 11.5% of the total protein for samples harvested in November to 4.5 and 6.3% for samples dug in August. These variations in free amino acid content of the raw material exceeded those induced by radiation and storage, even at levels as high as 4,500,000 rads. Steaming the irradiated unstored clams caused a slight additional increase in the free amino acid content, which was also higher in both air- and vacuum-packed clams irradiated with 350,000 and 450,000 rads and stored 30 days at 33°F than in raw unirradiated clams. It was not determined if the increase was due to the combined effects of irradiation and storage or to either one alone.

Data on the total amino acid content of the irradiated and control samples (Table II-3), although individual values varied somewhat, indicate that conditions of storage, heat processing, irradiation, or steaming did not cause significant changes in the total amino acids.

The various treatments did not cause any significant changes in the B vitamins (Table II-3). Niacin and vitamin B_{12} contents decreased slightly immediately after heat processing, but little further change in these vitamins occurred during 12 months of refrigerated storage. Clams irradiated at 4,500,000 rads and analyzed in the raw state showed an apparent decrease in riboflavin content, but this decrease was not evident in samples that were similarly treated but steamed for analysis. No significant differences were noted for vacuum packaging compared with air packaging.

The samples were clams packed in No. 2 C-enamel cans and (1) stored at 33°F with cans covered but not sealed, (2) heat processed for 20 min at 240°F, and (3) irradiated at levels of 350,000, 450,000, and 4,500,000 rads and stored at 33°F. Most of the amino acids were determined chromatographically.[27] Cystine was measured as cysteic acid by a slightly modified procedure of Schram, Moore, and Biguard.[28] Tryptophane was determined colorimetrically by the method by Bates,[29] later modified by Graham et al.[30] Riboflavin was determined by the method of McLaren, Cover, and Pearson[31] without extraction of the fat. Thiamin, niacin, pantothenic acid, vitamin B_{12}, and pyridoxin were determined by standard Association of Agricultural Chemists procedures and other standard methods.[32–34]

Research on Flavor and Odor

The flavors and odors of fish and shellfish become more pronounced during refrigerated storage, indicating that the compounds responsible for these attributes change in composition or concentration. Fishery products irradiated at pasteurization levels undergo somewhat similar changes in flavor and odor; higher irradiation levels usually cause more severe changes, making the product undesirable for eating. With detailed knowledge of the origin and fate of the

Table II-3 EFFECTS OF REFRIGERATED STORAGE, HEAT PROCESSING, AND IRRADIATION ON THE B VITAMIN, FREE AMINO ACID, AND TOTAL AMINO ACID CONTENTS OF CLAM MEATS

Treatment	Vitamin, μg/g												Free amino acid, % of total protein		Total amino acid, % of total protein		Moisture, %	
	Riboflavin		Thiamin		Niacin		Pyridoxin		Pantothenic acid		Vitamin B_{12}							
	Raw	Stmd.	Raw	Stmd.	Raw	Stmd.	Raw	Stmd.	Raw	Stmd.	Raw	Stmd.	Raw	Stmd.	Raw	Stmd.	Raw	Stmd.
Stored at 33°F																		
1 day	2.33	2.10	0.013	0.005	19.8	20.0	0.76	0.58	3.6	3.1	1.08	1.09	11.56	5.94	9.65	10.11	85.13	85.10
3 days	2.23	2.10	0.010	0.010	18.8	22.5	0.80	0.79	3.3	2.8	1.09	1.15	10.16	8.29	9.79	9.91	85.06	84.75
5 days	2.22	2.18	0.013	0.072	22.8	17.5	0.86	0.74	2.6	2.6	1.06	1.15	8.96	9.40	9.85	9.99	85.14	85.76
Heat processed, stored at room temperature																		
Control	1.91		0.016		25.3		0.80		3.10		1.05		4.46		12.04			
No storage		1.97		0.010		21.3		0.75		3.27		0.83		4.00		11.12		
6 months		1.95		0.030		18.4		1.02		3.12		0.71		3.56		12.03		
12 months		1.92		0.010		20.2		0.75		3.10		0.76		3.65		11.65		
Irradiated, stored at 33°F																		
Control	2.14		0.015		11.6		1.06		2.60		0.75		6.31		10.75		85.50	
4,500,000 rads air-packed																		
0 days	1.83	2.17	0.009	0.009	11.3	10.0	0.67	0.74	2.90	2.20	0.75	0.83	8.84	9.47	10.38	10.81	85.28	85.07
450,000 rads air-packed																		
30 days	2.12	2.07	0.012	0.009	9.8	9.1	0.67	0.74	3.00	3.40	0.69	0.66	9.49	9.43	8.97	9.19	88.28	87.83
350,000 rads vacuum-packed																		
30 days	2.37	2.13	0.010	0.010	11.2	11.3	0.99	0.80	3.00	3.00	0.68	0.68	10.90	10.08	10.05	10.29	86.30	86.20

flavor and odor compounds in fish, it may be possible to take steps that will inhibit the changes. This could permit the use of radiation preservation for fishery products not now amenable to this process, and, for those which are, it might permit use of higher dose levels and greater extension in refrigerated shelf life. Our research was designed to establish differences between the changes in volatile compounds of irradiated and nonirradiated fish, investigate methods of measuring these changes and correlating them with the quality of irradiated and nonirradiated products, and identify the components principally responsible for flavor and odor changes in fishery products. Our findings to date indicate that irradiation at levels of 350,000 to 450,000 rads, cooking, and extended refrigerated storage increase the number, and in some cases the concentration, of the volatile compounds in clam meats but that combinations of these processes do not have an additive effect. A typical chromatogram of the head gas from the total condensate of distilled clam meat (Fig. II-6*a*), although the data are limited, indicates that at least nine different volatile compounds were present. For convenience the chromatogram peaks are labeled with letters. The effluent gas of peak A had a distinct odor of hydrogen sulfide. Peak D sometimes (though not in the chromatogram shown in Fig. II-6*a*) has two components; peak E represents the largest single concentrate of head-gas volatiles. Odor and retention time indicate that this compound was dimethyl sulfide. Recently, emphasis has been placed on analyzing the carbonyl compounds from the total condensates. A typical gas chromatogram of the carbonyl compounds from volatiles of irradiated vacuum-packed clam meats is shown in Fig. II-6*b*.

The compounds studied were removed from the fish by a high-vacuum low-temperature distillation technique described previously by Merritt.[35] Separation was performed on a gas chromatograph fitted with a ^{90}Sr ionization detector and an attachment that programs column temperature from −65 to +65 °C. The column was acid-washed Chromosorb W coated with 2% oxydipropionitrile.

The samples were haddock fillets and clam meats separately packed in No. 2 C-enamel cans and either (1) sealed under a vacuum of 27 in. Hg or air-packed at atmospheric pressure or (2) irradiated at levels of 50,000 to 550,000 rads and stored at 34°F for 45 days for the fish and 30 days for the clams. At intervals, samples were removed from refrigerated storage, and total volatiles were collected.

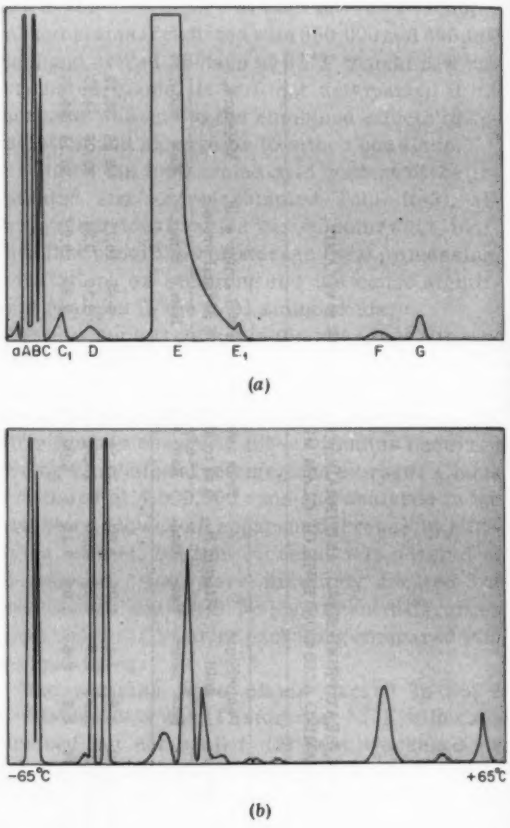

Fig. II-6 Typical gas chromatograms of (*a*) total condensate head gas and (*b*) volatile carbonyl compounds in clam meats.

As our research on flavor and odor proceeds, additional techniques for collecting, concentrating, and regenerating the volatile compounds in fish will be studied. Selected column packings will be used to permit more efficient separation; a mass spectrometer will be used to identify the compounds in the test samples.

Optimum Levels of Radiation for Fish and Shellfish

Several series of investigations were carried out to determine for each of several species of fish and shellfish the dose that provided a maximum extension of shelf life and a minimum change in product acceptability, as determined by a taste panel.

The species studied were soft-shell clam meats and fillets from haddock, pollock, and ocean perch. Flavor and odor were the principal factors considered in evaluating product quality. Although the texture and appearance were also judged, they were of less importance. The optimum radiation dose (Table II-4) varied somewhat with the individual product; for example, soft-shell clam meats could be irradiated to a higher dosage than fillets without adverse changes in flavor and odor. Haddock appeared to respond to irradiation more favorably than ocean perch or pollock. Irradiated products stored at 42°F had a much shorter shelf life than those stored at 33°F. In general, when the fillets were deep-fat-fried they were of greater acceptability and could be subjected to higher doses without noticeable changes in flavor and odor. The storage times shown are based on an average of the results of taste-panel tests conducted on steamed fillets and deep-fat-fried clam meats.

All samples were of known history and in most cases were only 1 day out of the water when irradiated. They were packed in No. 2 C-enamel cans, under air or under a vacuum of 27 in. Hg, and were irradiated at levels from 50,000 to 550,000 rads. After irradiation, samples were stored at the laboratory at 33 and 42°F. At intervals, cans were removed from the chill room, and the product was cooked and served to the taste panel. Difference and acceptability taste-panel tests were carried out for each product.

Difference tests were done in duplicate for each species. At each sitting of the panel, five samples were evaluated. Four of these were irradiated at different levels, and the fifth was an unirradiated control. Control fillets were fresh frozen, and control clams were freshly shucked. All samples were identified only by code. Fillets were steamed, and clams were always deep-fat-fried. The taste panel scored the samples for appearance, odor, flavor, and texture on a 5-point scale ranging from excellent (=5) to poor (=1).

In a separate series, acceptability tests were carried out in which a 9-point hedonic scale ranging from "like extremely" (=9) to "dislike

Table II-4 OPTIMUM RADIATION LEVELS AND STORAGE TIME FOR FISH AND SHELLFISH

Product	Radiation level, rads		Score†		
	Used	Optimum*	30 days' storage, 33°F	10 days' storage, 42°F	20 days' storage, 42°F
Soft-shell clam meats	50,000 150,000 250,000 350,000 450,000	450,000	3.3	3.8	
Haddock fillets	50,000 150,000 250,000 350,000	250,000	2.9		2.7
Ocean perch fillets	50,000 150,000 250,000 350,000	250,000	2.4		2.2
Pollock fillets	50,000 150,000 250,000 350,000	150,000	2.0	2.5	

*Often, for radiation levels lower than optimum, the shelf life at 33°F was less than 30 days; levels higher than optimum usually resulted in slightly lower scores because of the appearance of radiation-induced off-flavors. As would be expected, scores from the panel tests indicated clearly that 42°F was an unsatisfactory storage temperature since (1) spoilage occurred more rapidly than at 33°F and (2) the quality level during any storage period prior to spoilage was lower than at 33°F.

†Score based on difference tests; 5 = excellent, 1 = poor.

extremely" (= 1). All samples were deep-fat-fried and were presented to the panel by the single stimulus method.

Effect of Preirradiation Quality on Postirradiation Shelf Life

The extension of product shelf life reported previously is for samples that were 1 day out of the water when irradiated. This condition is seldom realized in commercial practice where fish available for irradiation might be 5 to 8 days old and of varying quality. The results of three experiments, conducted to determine the commercial feasibility of this process when applied to fish of high to low quality, are not conclusive but indicate that fish should be held in ice for no longer than 7 to 9 days prior to irradiation (Fig. II-7). When the raw material is held for longer periods of time, the shelf life of the irradiated product is greatly shortened. These findings also indicate that there was considerable variation in the quality of the fresh haddock obtained for the laboratory studies. For example, the postirradiation shelf life of the 7-day-old iced fish varied from 23 to 33 days.

For these tests approximately 600 lb of fresh eviscerated haddock was used in each experiment. The fish were packed in boxes with ice, and, at 2- to 3-day intervals, samples were removed, filleted, skinned, and air-packed in No. 2 C-enamel cans. They were then irradiated at a level of 250,000 rads. The irradiated samples were stored in the laboratory at 33 to 35°F. At 3- to 5-day intervals, the fillets were removed from refrigerated storage, steamed, and served to the taste panel together with a control of frozen fillets prepared from iced haddock 1 day old.

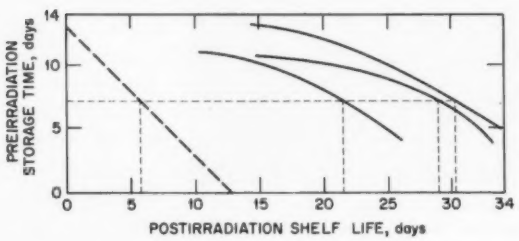

Fig. II-7 Postirradiation shelf life at 35°F of eviscerated haddock as a function of preirradiation storage time on ice. - - -, not irradiated; ——, 250,000 rads irradiation (three runs).

In the absence of objective methods of measuring quality changes, it is rather difficult to define precisely the extension of shelf life that can be realized when irradiating iced fish of varying quality. It should be emphasized, however, that these results are preliminary, and more extensive investigations are now being conducted in which microbiological analyses are being used to assist in defining product quality.

Summary

Studies on the application of pasteurization levels of radiation to certain species of New England fish and shellfish are being conducted at the Bureau of Commercial Fisheries Technological Laboratory in Gloucester, Mass. The results to date show that the amino acids and B vitamins of clam meats are not adversely affected by irradiation at levels as high as 4,500,000 rads. Several compounds have been identified in clam meats from known retention times in gas-chromatography columns. Mass spectrometry is being used to identify more of the many compounds that make up the flavor and odor of these products. Organoleptic studies on irradiated samples of soft-shell clam meats and fillets of haddock, ocean perch, and pollock indicate that, when supplemented with storage at 33°F, this technique will triple the product shelf life without adverse changes in flavor and odor. In general, these products can be irradiated at levels less than 500,000 rads and stored for 30 days at 33°F. Storage at 42°F results in about a 50% decrease in the shelf life of the irradiated seafood. The quality of the raw material significantly influences the shelf life and acceptability of the irradiated product. Fish stored longer than 7 to 9 days in ice should not be irradiated because of the limited shelf life of the product.

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Section

III

Isotopes and Radiation Technology

Isotopic Power Development

Editor's Note: Since the beginning of the Systems for Nuclear Auxiliary Power (SNAP) program in 1955, a number of new problems have evolved which are of concern to the U.S. Atomic Energy Commission (AEC) and other government agencies charged with the support of this effort. These problems are concerned with the status of requirements, the magnitude and scope of the SNAP program, the long-range effort, the development of alternative, nonnuclear power devices, and to some extent the interrelation between the AEC program and that of the ultimate users, the National Aeronautics and Space Administration and the Department of Defense.

Recognizing that the interplay of technical and programmatic changes is greatly influencing the SNAP program, the Chairman of the AEC announced in November 1962 that the Commission had decided to take a new look at the SNAP program and directed the staff of the Office of Program Evaluation of the Division of Reactor Development to undertake this study. The evaluation of the SNAP program by this group was recently published,[1] and the Commission then published its report[2] based on this evaluation. The introduction and conclusions of the Commission's report, which are reproduced here with minor editorial changes, outline the official philosophy and scope of the AEC's future program in the SNAP program. The complete SNAP Isotope Program section of the evaluation report[1] is also included since it contains much that is important to isotope producers and users; however, the preface to this evaluation emphasizes that it does not represent an official position of the AEC on any of the subjects covered.

AEC Report on Systems for Nuclear Auxiliary Power (SNAP)[2]

Man, as he moves into the space age, will encounter many unique and difficult problems, not the least of which will be his ability to produce the power necessary to accomplish his ambitious objectives.

The energy that can be obtained from the atomic nucleus is an advantageous and vitally needed source of space power. The responsibility of the Commission in such a situation was clearly stated by Congress. The policy of the United States is that development, use, and control of nuclear energy shall be directed by the Commission so as to make the maximum contribution to the general welfare, subject at all times to the paramount objective of making the maximum contribution to the common defense and security; it is the stated purpose of the Atomic Energy Program to provide for conducting, assisting, and fostering research and development in order to encourage maximum scientific and industrial progress.

The AEC is unique among Executive Branch departments and agencies in having for its major purpose the development of as specific a field as nuclear energy. Other agencies of the Executive Branch have broadly defined scientific, technical, and operational responsibilities for contributing to the general welfare and security of the nation. Having a more specific responsibility, the Commission does not retain sole authority and responsibility over nuclear appli-

cations as developmental projects approach completion and as nuclear devices start to meet specific power requirements and thus enter the mainstreams of national activity. To an increasing extent the authority then spreads to others; thus successful developments in nuclear auxiliary power for space will be followed by Department of Defense (DOD) and National Aeronautics and Space Administration (NASA) procurement of the demonstrated systems. At such times the continuing interests of the Commission emphasize assuring the continued safety of the general public and the availability of supplies of fuel adequate for users' needs.

In keeping with its responsibilities under the Act, the AEC has engaged in vigorous development and demonstration of nuclear power systems for space missions of interest to NASA and DOD. The total effort, which also involves participation by other agencies, includes development of rockets that use nuclear reactors to provide heat energy to propellant gas and of systems for nuclear auxiliary power. The latter, called "SNAP units," can provide electric power for such uses as operating sensing devices; transmitting and receiving radio, radar, and television signals; providing the heat and electric power needed to support astronauts for long periods of time; and ultimately providing the means to propel rockets to greater distances at higher speeds by electrical acceleration of the rocket exhaust. A summary of SNAP generators is given in Tables III-1 and III-2.

All SNAP units generate heat energy, part of which can be used to make electricity. Radioisotopic SNAP units stay at a high temperature for years due to a slow process of spontaneous radioactive change going on within the units. Electrical circuits of thermoelectric materials, when placed tightly against the hot outer surface of the radioisotope containers, generate a flow of electricity between hot and cold parts of the circuit. This electricity is used to operate the payload. Reactor SNAP units obtain their energy from neutron-induced fission of uranium. SNAP reactors differ from other reactors primarily in their compactness: typical reactor cores are contained in vessels about

Table III-1 RADIOISOTOPIC SNAP GENERATORS

Designation	Use	Power, watts*	Weight, lb	Isotope	Design life	Current status
			Space Applications			
SNAP-3	Demonstration device	2.5	4	^{210}Po	90 days	Program completed
Undesignated (modified SNAP-3)	Navy navigational satellites	2.7	4.6	^{238}Pu	5 years	Generators launched June and November 1961; first still in operation; second failed after 8 months
SNAP-9A	Department of Defense satellites	25	27	^{238}Pu	5 years	Generators launched September and December 1963; both in operation
SNAP-11	Moon probe	21–25	30	^{242}Cm	90-day mission	Being tested
SNAP-13	Thermionic demonstration device	12	4	^{242}Cm	90-day mission	Being fabricated
SNAP-17	Communications satellite	25	28	^{90}Sr	5 years	Initiated in mid-1963
SNAP-19	IMP, Nimbus	20	18	^{238}Pu	5 years	Design-study stage
Undesignated	Lightweight demonstration device	6–10	3	^{238}Pu or ^{90}Sr	Greater than 1 year	Being tested
			Terrestrial Applications			
Undesignated	Axel Heiberg weather station	5	1680	^{90}Sr	2 years minimum	Operating since August 1961
SNAP-7A through 7E	Navigational buoys, lights and beacons, weather stations, etc.	6.5–60	1900–6000	^{90}Sr	10 years	First unit in operation February 1962; four now operating
SNAP-15A and 15B	Nuclear weapons	0.001	1	^{238}Pu	4 years	Being tested
SNAP-21	Ocean bottom	10		^{90}Sr	5 years	Initiated in late 1963

*All power ratings in this section are in electrical watts or kilowatts.

Table III-2 SPACE REACTOR SNAP GENERATORS

Designation	Use	Power, kw Design	Power, kw Potential	Weight,* lb	Design life	Current status
			Reactor Projects			
SNAP-10A	Thermoelectric demonstration system	0.5	2	1000	1 year	Nearing completion
SNAP-2	Rankine-cycle demonstration system	3	10	1470	1 year	Reoriented toward component improvement
SNAP-8	Communication satellites or space station	35		6000 (est.)	10,000 hr	Power test of experimental reactor now under way
SNAP-50	Electric propulsion and auxiliary power	300	1200	6000 (est.)	10,000 hr	Development of components continuing; construction of reactor experiment terminated
			Research and Development Programs			
Hydride reactors						
With thermoelectric power conversion			15	3600		
With turboelectric power conversion			100	5000		
Boiling potassium reactors			140–400			
Refractory metals reactor						

*Includes weight of a radiation shield to reduce cumulative dose over a year's operation to levels that will not damage sensitive instrumentation.

the size of a 5-gal can. Reactor heat is transported by a fluid coolant from the core to equipment for converting heat to electrical energy.

Terrestrial applications of isotopic SNAP systems and technology are to be expected and have been made. These devices have demonstrated their unique advantages for use in rigorous environments and in barren, inaccessible places. Automatic weather stations in polar regions and at sea, navigational signals, and ocean-bottom beacons have been developed and are being demonstrated in the terrestrial SNAP program.

The program to develop SNAP units includes research and the engineering development needed for proof of technical feasibility and safety of the more promising types of systems, as well as project-oriented efforts to demonstrate the practical feasibility and utility of specific systems. The program has taken full advantage of the extensive useful information and the experienced scientists and engineers available from other established military and civilian programs. It has achieved notable success in developing small systems powered by radioisotopes. The program to develop and demonstrate larger systems powered by nuclear reactors is well under way but will require much more time and effort and more extensive facilities than the earlier efforts on smaller systems.

In order to consider the magnitude and timing of the need for systems for nuclear auxiliary power, especially in meeting present and future space power demands, this report* first considers the magnitude, characteristics, and likelihood of occurrence of uses for all auxiliary power systems and then discusses the extent to which nuclear, or other, power systems can meet them. This report also considers the programs under way and planned for the future and finally presents the Commission's conclusions on the overall SNAP program.

Conclusions

The SNAP program was begun as a result of early recognition of anticipated electrical power needs for spacecraft. The AEC's program has pioneered in the applications of nuclear power for this purpose and has been primarily responsible, in government, for many novel approaches. Such things as compact reactors, radioisotope heat-energy capsules, thermoelec-

*The items enumerated here, with the exception of the conclusions, are not included in this excerpt but may be seen in the full report, Ref. 2.

tric generators, thermionic generators, compact mercury-vapor turbines connected directly to electric generators, light-weight shielding, and aerospace safety are but a part of the developments that the Commission has been pursuing in support of the nation's space program.

This SNAP study has confirmed previous statements concerning the advantages of and needs for nuclear power in space. With the passage of time and as the technology has been carried closer to flight applications, some developments appear to be more difficult and require more time. On the requirements side, time schedules have slipped, especially for missions for which nuclear power is necessary. Since one of the most expensive phases of any project-oriented SNAP development is in flight testing, it is important that such testing be done at a time which will make maximum contribution toward answering the uncertainties, especially technological uncertainties of operation in the space environment.

The present AEC annual cost for the SNAP research and development effort, including directly related efforts in isotope development and aerospace safety, is approximately $100 million. Continued operation at approximately this level should permit development of the technology of isotopic and hydride reactor systems with their associated converters to a state where specific systems satisfying requirements in the range up to tens of kilowatts* could be assembled for various mission tests by the late sixties. This level of effort does not provide for flight testing. Increased work necessary for project-oriented systems would require additional support for the extensive systems-engineering development and flight testing needed to meet specific mission requirements. The present program of advanced technology and its associated ground testing is proceeding at a relatively low but reasonable level consistent with long-term development. It must be recognized that full development of an advanced system to provide power in the range of hundreds to thousands of kilowatts will in itself probably cost over a billion dollars during a period of 15 or more years.

This study has led to several conclusions which have formed the basis for program planning including several actions taken by the Commission during 1963. These several conclusions are set forth below.

1. *Nuclear auxiliary power offers unique advantages to a wide variety of space missions. The SNAP development program should be concentrated on approaches to provide these advantages, which include among others, light-weight, compact fission and radioisotopic systems in a power range up to 20 kilowatts and reactor systems with long life at higher power.*

In certain space applications, electrical power requirements can be met only by nuclear systems. In others, there may be a competition among chemical, solar, and nuclear systems, and the user will have to weigh the advantages and disadvantages, both technical and economical, in making a selection for specific missions. The Commission's research and development program on nuclear technology must be directed at exploiting advantages such as long life, high power, compactness, and ruggedness and at reducing the present disadvantages of shield weight, high cost, and radiation emission. The development of the technology and the testing of systems must be carried sufficiently far that users' judgments in the selection of systems will be based upon firm technological grounds.

2. *The performance of ambitious space missions will require amounts of reliable power so large that they can be achieved only from nuclear systems. Continued timely development of advanced SNAP technology is necessary to meet this need.*

Advanced technology is required in the development of minimum-weight systems in the power range of hundreds to thousands of kilowatts. Although the mission requirement is far in the future, the development of the technology will require a long lead time.

3. *In the absence of near-term specific mission requirements, the SNAP reactor technology must be developed to meet power requirements within several power ranges (kilowatts, tens of kilowatts, and hundreds to thousands of kilowatts), thereby making available a technological base for the specific systems for ultimate mission use.*

The grouping of potential requirements into several categories, each of which differs from

*All the power ratings used in this section are in electrical watts or kilowatts.

the other by an order of magnitude, will permit reactor development and converter development to go forward to satisfy the needs set forth above with a minimum number of system concepts. Obviously, within these ranges, developments will be directed toward those potential applications in which users indicate the greatest interest. Potential users must recognize that adequate time must be allowed in their own programs for the development of a suitable nuclear system. However, the Commission's program should assist in minimizing the time necessary to assemble a specific system for ground and flight test.

4. *Development of SNAP technology should be carried forward with the Commission giving full attention to the integration of the converter with the nuclear heat source.*

The user is most concerned that equipment be available to meet this electrical power requirement. In a program of advancing technology directed toward meeting these electrical power needs, the nuclear heat source–converter system must be developed in a coordinated manner. The Commission must assure itself that adequate development is going forward on both components and take the initiative in meeting the technological problems. It is recognized that other agencies, as well as the AEC, will be involved in development of converter technology. Also, when specific development projects are undertaken on a reasonably well defined time schedule, program responsibilities will be determined jointly with the requesting agency.

5. *Where Commission responsibility for power system developments involves first-of-a-kind technology and equipment, the program should extend through the demonstration phase, including ground and flight testing as necessary, and should encompass programs for the resolution of safety problems.*

First-of-a-kind technology for nuclear heat source–converter systems should be carried through a demonstration phase which may, in addition to ground testing, require flight testing. Such a program will give the Commission better guidance for its future work and will provide more extensive information for the user on which he can base future plans. When flight launch and tracking facilities are required to meet the needs of the Commission's program, the Commission would plan for and arrange with NASA or DOD to carry out the flight. Where joint projects are involved, the Commission would expect the cooperating agency to plan for and execute the flight test. The Commission's program will benefit from participation with users in planning and carrying out launch operations, assuring safety, and finally testing of the SNAP device in space.

6. *The orderly development of SNAP technology through the demonstration phase should provide users with information and experience needed for them to specify and obtain nuclear power systems and to assist in planning their missions.*

In selecting a power supply to meet requirements, user agencies must have available sound technical and economic data and, to the extent possible, performance experience at or near ultimate operating conditions. In implementation, the Commission's program will approach such needs, while deferring the incurrence of major project expenditures until such times as specific needs can be set forth.

The SNAP program has reached a point in technology where its importance to the nation must be recognized, accepted, and supported, or the development and performance of ambitious space missions will be handicapped by a lack of adequate and reliable power.

SNAP Isotope Program

Requirements and Potential Applications for Isotopic SNAP

Recent compilations by NASA and DOD of probable space electric auxiliary power needs regardless of the type of power generator to be employed, i.e., solar cell, isotopic, or battery for unmanned satellites and probes during 1964–1967 indicate that about 20,000 electrical watts in total might be required. The details are given in Table III-3. Mission schedules beyond 1967 are a matter of speculation; however, a very rough estimate is that the total annual requirement for power generators of less than 1000 watts' capacity will probably be in excess of 5000 watts/year.

Of the various space missions, several are of a system character: the weather satellite system (Tiros and Nimbus), the communica-

Table III-3 SPACE ELECTRIC POWER REQUIREMENTS—TENTATIVE FLIGHT SCHEDULE

Mission	Approx. power at steady load, watts	Number of flights			
		1964	1965	1966	1967
Scientific Satellites and Probes					
Astronomy					
Orbiting astronomical observatories	500		1	2	1
Solar physics					
Orbiting solar observatories	25	2	1	2	1
Advanced orbiting solar observatories	15–50				2
Geophysics and Geodesy					
Orbiting geophysical observatories	500	1	3	1	3
Explorers	15–50	1	3	1	2
Monitors—Interplanetary Monitoring Probe	40	1	3	2	2
International	15–50	7	5	6	4
Anna Geodetic Satellite	50		3	2	1
Biophysics					
Biological satellites	25		1	4	1
Lunar science					
Ranger—lunar impact	150–190	4			
Surveyor Lander	15–25*		5	2	
Surveyor Orbiter	125			4	3
Planetary and interplanetary science					
Mariner (Mars and Venus)	150–250	2		4	2
Pioneer	5–100		3	3	1
Satellite Applications—Nondefense					
Meteorology					
Tiros	25–50	1			
Nimbus	140–250	1	1	1	1
Communications satellites					
Telstar	60	1	12	10	8
Relay	50	2			
Syncom	25–100	1	1	2	
Satellite Applications—Military Defense					
Communications satellites					
Medium altitude	25				
Syncom	100				
Navy navigational satellites	25				

*Power when landed; 100 watts en route.

tions satellite system (Telstar, Relay, and Syncom), the military-communications satellite system, and the Navy navigational satellite system. Such systems are usually maintained by periodic replacement of orbiting satellites that have either failed or reached the end of their useful life. Satellites making up these four systems account for about one-third of the total electric power requirements during the 1964–1967 period.

Isotopic power sources will be used in some of the space missions shown in Table III-3, but the total number of such applications is unknown at this time. Isotopic power generators have been developed for the Navy navigational satellites (SNAP-9A), the Surveyor Lander (SNAP-11), and the Interplanetary Monitoring Probe (IMP generators). Whether the latter two applications will take place is yet uncertain. Work on several designs of a 25- to 50-watt strontium generator suitable for a medium-altitude military-communications satellite has been initiated. NASA has inquired into the feasibility of using isotopic power on the orbiting astronomical observatories and Nimbus weather satellites. Other missions that could quite conceivably employ radioisotope thermoelectric generators are the Anna Geodetic Satellite, Surveyor Orbiter, and the communication satellites Telstar, Relay, and Syncom. Radioisotope thermoelectric generators could be used on geophysical and biophysical satellites

if they do not interfere with the various onboard radiation-measuring experiments. The solar observatories are sun oriented and therefore will most likely use oriented solar-cell systems.

Auxiliary electric power needs for manned space missions have not been included in this compilation, since it was felt that they would be definitely scheduled by firm requirements if the user agencies desired to use isotope generators in any of these flights.

Potential large-scale terrestrial applications of isotopic power sources (Table III-4) are under consideration and could require more than 20,000 watts of generator power for the 1964–1967 period.

Isotope Availability and Cost

Isotopes that are currently being produced as power sources for space radioisotope thermoelectric generators (RTG) are ^{238}Pu for SNAP-9A and ^{242}Cm for SNAP-11 and -13. Strontium-90 is used as a power source in the SNAP-7 series terrestrial generators.

The AEC has experience in producing ^{238}Pu and plans to make 30 to 35 kg available prior to 1968, which at a 5% overall RTG efficiency, would provide about 900 watts of power. Thereafter, estimated annual availability is equivalent to about 550 watts by the end of 1968, increasing to about 800 watts by the end of 1972. For planning purposes, these availability figures roughly represent a lower limit to the amount of ^{238}Pu that could be produced by the AEC at a cost of about $450 per gram ($16,000 per watt). The upper limit on availability at this cost is not likely to exceed twice the amounts stated above. Alternatives for substantially increasing ^{238}Pu availability are concomitant with unit costs of several thousand dollars per gram. Hence, expanding AEC production of ^{238}Pu with its concomitantly higher unit cost may be undesirable, unless the user agencies specifically request additional amounts of this isotope in spite of its cost.

Radioisotopes which have possible use as heat sources and which would be available during the next several years, in addition to ^{238}Pu, are ^{90}Sr, ^{137}Cs, ^{147}Pm, ^{244}Cm, ^{242}Cm, ^{210}Po, ^{144}Ce, and ^{60}Co (Table III-5). The anticipated annual availabilities and unit costs during the 1964–1966 period are given in the following tabulation.

Isotope	Annual capacity	Unit cost
^{90}Sr	3–5 Mc	$1.00–1.50/curie
^{137}Cs	1–3.5 Mc	$0.80/curie
^{147}Pm	0.3 Mc	$0.75/curie
^{242}Cm	150 g	$12,700/g

Again, assuming a 5% overall RTG efficiency in converting isotopic heat output to usable electric power, the thermal output from the above isotopes at the time of separation and purification might provide the following amounts of electric power:

Isotope	Annual capacity	Unit cost
^{90}Sr	1000–1600 watts	$3000–4500/watt
^{137}Cs	250–850 watts	$3500/watt
^{147}Pm	6 watts	$40,000/watt
^{242}Cm	900 watts	$2000/watt

The short half-life of ^{242}Cm (163 days) radically affects the cost of power. For example, the cost of providing 1 electrical watt at the end of a 1-year mission would be $10,000, or roughly five times the cost of power from freshly separated ^{242}Cm. Because of its rapid decay, ^{242}Cm is not considered a candidate for terrestrial applications. The ^{90}Sr half-life of 28 years, relatively low cost, and sufficiently high power density from various compounds makes it a feasible RTG fuel for either space or terrestrial applications. The low power density of suitable ^{137}Cs compounds, plus the fact that a large fraction of the heat energy must be recovered from gamma radiation, limits its use to terrestrial applications. The cost of ^{238}Pu, relative to ^{90}Sr or ^{137}Cs, virtually precludes its use from other than very special terrestrial systems. Because of its low radiation background, ^{147}Pm, like ^{238}Pu, is an attractive candidate for space applications. Several other fuels, e.g., ^{144}Ce, ^{210}Po, ^{170}Tm, ^{60}Co, ^{232}U, ^{228}Th, and mixed fission products, have also been considered as possible heat sources. However, they are generally marginal compared with the other isotopes discussed above from the standpoints of production feasibility, properties, and cost.

When isotope availability from present facilities is compared with potential space requirements, it is apparent that the electric power from ^{238}Pu that could be made available is far

Table III-4 POTENTIAL TERRESTRIAL APPLICATIONS OF ISOTOPIC SNAP UNITS

	Approx. generator power, watts	Potential 1964–1967 requirements	
		No. of generators	Total watts
Military Defense Applications			
Navigational aids — Navy	10		
Antisubmarine warfare coastal surveillance — Navy	150		
Oceanographic and weather monitors — ASW support — Navy	20		
Dormant missile system — Air Force	100		
Government and Civilian Nondefense Applications			
Weather stations — interagency	20–60	50	1000–3000
Navigational aids			
Alaskan and Aleutian shore lights — Coast Guard	5–15	70	350–1050
Buoys — Alaskan waters — Coast Guard	10	70	700
Gulf of Mexico oil wells — Civilian	50	100	5000
Beacons — Coast Guard	5–100	20	100–2000
Unattended seismic stations — interagency	60	10–100	600–6000
Missile-range underwater splash detectors and tracking-ship position fixers — Air Force	10	260	2600

Table III-5 CHARACTERISTICS OF ISOTOPIC HEAT SOURCES[3]

	^{90}Sr	^{137}Cs	^{147}Pm	^{238}Pu	^{244}Cm	^{242}Cm	^{210}Po	^{144}Ce	^{60}Co
Type of decay	Beta	Beta-gamma	Beta	Alpha	Alpha	Alpha	Alpha	Beta-gamma	Beta-gamma
Half-life, years	28	30	2.7	89	18	0.45	0.38	0.78	5.3
Specific power of isotope, kw/g	0.90	0.42	0.33	0.56	2.8	120	141	25.6	17.4
Estimated isotopic purity, %	50	35	95	80	98	90	95	18	10
Typical fuel form	SrO	Glass	Pm_2O_3	PuO_2	Cm_2O_3	Cm_2O_3	Metal	CeO_2	Metal
Active isotope in compound, %	42	16	82	71	89	82	95	15	10
Specific power of compound, kw/g	0.38	0.067	0.27	0.39	2.49	98	134	3.8	1.7
Density of compound, g/cm^3	3.7	3.2	6.6	8.9	10.6	11.75	9.3	6.4	8.9
Power density of compound, kw/cm^3	1.40	0.21	1.8	3.5	26.4	1150	1210	24.5	15.5
Shielding requirement	Heavy	Heavy	Minor	Minor	Moderate	Minor	Minor	Heavy	Heavy
Emission requiring shielding	Bremsstrahlung	Gamma			Neutron			Gamma	Gamma

short of possible space power requirements. This situation is not significantly affected by either the ^{147}Pm or ^{242}Cm that might be available from present facilities. The 3 to 5 Mc per year of ^{90}Sr from existing facilities would provide about 1000 to 1600 watts of power. However, its future large-scale use in space is as yet undetermined. This is discussed below under safety considerations.

There are essentially two alternatives by which the AEC could substantially increase the availability of isotopes suitable for use as heat sources. One is large-scale separation, purification, and encapsulation of radioactive fission products in the Hanford wastes and the other is ^{244}Cm production. However, neither of these would aid in the availability of isotopes prior to 1967 or 1968.

The Commission is proceeding with plans for a fission-product recovery, purification, chemical conversion, and encapsulation facility at Hanford. The processing capacity for this facility is yet undecided, but it is not unreasonable to assume that it will be about:

Isotope	Annual capacity, Mc	Unit cost
^{90}Sr	10	10–15¢/curie
^{137}Cs	10	6¢/curie
^{144}Ce	100	1¢/curie
^{147}Pm*	15	7–10¢/curie

*Aged 1 half-life.

The above unit costs are, at best, rough estimates and serve merely to indicate what might be expected from such a facility if operated at its rated capacity. Assuming a 5% overall RTG efficiency, the output of ^{90}Sr might provide in excess of 3000 watts of power per year; ^{137}Cs, about 2500 watts; and, at the time of separation, 270 watts from the ^{147}Pm and 40,000 watts from the ^{144}Ce (although its short half-life of 284 days introduces power flattening problems in RTG applications).

Curium-244 is of particular interest because of its extremely desirable characteristics for SNAP applications, notably an 18-year half-life and a specific power more than three times greater than any other suitable long-lived isotopes. Because of its specific power, ^{244}Cm could be used in conjunction with thermionic converters, which are expected to be roughly twice as efficient in converting heat to electricity as thermoelectric converters, thereby optimizing the use of a valuable and limited resource. A study of large-scale ^{244}Cm production indicates that significant outputs of relatively pure ^{244}Cm could be produced at a cost in the range of $1000 to $2000 per gram. Several years of in-pile irradiation, out-of-pile cooling, and reprocessing are required in producing curium of high purity in the 244 isotope. The annual output during the first few years thereafter might be between 20 and 30 kg and could provide roughly 6000 to 8000 watts in thermionic generators operating at 10% overall efficiency or 3000 to 4000 watts of power per year in RTG's operating at 5% overall efficiency.

A program at Savannah River to produce 3 kg of ^{244}Cm was begun in the spring of 1964. This will provide production and separations experience as well as the necessary material to demonstrate the feasibility of using ^{244}Cm for space applications.

Isotopic SNAP Safety and Radiation Considerations

Once an isotope generator has successfully been placed in a long-lived (more than 200 years) orbit, the isotope will decay to an insignificant activity level prior to returning to the earth. However, earlier random reentry of highly active isotopes could take place in the event of failure to achieve the intended long-lived orbit after high velocities had been reached. The question that arises is: Should capsules be designed to burn up or to return intact? Since burnup adds to radioactive fallout, it becomes necessary to evaluate such things as (1) the amount of ^{90}Sr, ^{238}Pu, and ^{244}Cm which would be introduced as fallout and which could be safely tolerated, (2) the fallout particle size, and (3) the distribution of fallout. Some idea of the magnitude of this problem may be gleaned from the following: the yield from a 10-megaton fission produces approximately 1 Mc of ^{90}Sr. The approximate fission yields of nuclear weapons tests conducted in the atmosphere by all nations through 1962 produced about 19 Mc of ^{90}Sr. A single 100-watt RTG fueled with ^{90}Sr would contain about $\frac{1}{3}$ Mc, which, if burned up due to a faulty or short-lived orbit, would be equivalent to that produced by a 3-megaton fission yield. With regard to ^{238}Pu, one SNAP-9A, if burned up after early return, would add a

significant fraction to the 38,000 curies of ^{238}Pu and 164,000 curies of ^{239}Pu activity estimated to be in the stratosphere as of mid-1963. Moreover, the maximum permissible concentration of ^{238}Pu in air for the general population is 150 to 200 times less than that of ^{90}Sr, and ^{244}Cm is 30 to 70 times less than ^{90}Sr.

Capsules not burned up might return on land. Recent calculations indicate that the radiation dose at 1 ft from a relatively unaged encapsulated fuel source designed for a 10- and a 100-watt RTG might be:

Isotope	Rems per hour at 1 ft*	
	10 watts	100 watts
^{238}Pu	0.2	2.0
^{147}Pm	0.25	2.5
^{244}Cm	6	60
^{90}Sr	325	1650

*Equivalent of 1 cm of lead assumed for attenuation of very soft gamma.

The ^{90}Sr and ^{244}Cm dose rates are such that it appears necessary to either recover the capsules promptly or assure that they do not return on land if returned intact. This would require either very accurate tracking, an independent system for controlled reentry, or injection into outer space.

Promethium-147 and perhaps ^{238}Pu could return intact from short-lived or faulty orbits and, because of their very low radiation, might not necessarily need to be deliberately recovered.

The above dose rates also present a fuel-handling problem at the launching site. A generator loaded with ^{244}Cm would require some shielding on the gantry if personnel were to work nearby; a ^{90}Sr-fueled generator most certainly would. It might be preferable to load encapsulated ^{244}Cm into a generator either remotely or with tongs just prior to launching and thereby avoid the problems concomitant with shielding and personnel radiation monitoring. Encapsulated ^{90}Sr sources require remote loading just prior to launching. Remote handling on the missile gantry introduces complications that are not shared by ^{238}Pu and ^{147}Pm due to their low radiation backgrounds.

Generators fueled with ^{244}Cm or ^{90}Sr require either shielding or separation from the spacecraft to avoid long-term radiation damage to vulnerable payload components such as germanium and silicon semiconductors. Shielding the payload can impose a severe penalty on the total weight associated with the auxiliary power system, depending on the source strength, geometry, and distance from the dose point. This can be avoided by physically separating the isotopic generator from the electronics with a boom. The boom lengths required are roughly:

Generator power, watts	Boom length, ft*	
	^{90}Sr	^{244}Cm
25	7	5
100	12	11
250	16	17
1000	30	34

*Separation of isotopic source and payload needed to confine integrated dose to 10^6 rads and 10^{12} neutrons/cm^2 in 5 years with no shielding.

Booms might be in a folded attitude during launching and deployed after injection of the spacecraft into orbit. The Navy navigational satellites use a 60-ft boom; the NASA orbiting geophysical observatories will have various experiments requiring preferred orientations or isolation mounted on external booms, etc., so that booms are not uncommon to today's spacecraft.

Economics of Isotopic SNAP

Economics apparently are currently a major consideration in the various nondefense SNAP terrestrial applications, especially in cases such as replacing batteries with isotopes in buoys and remote shore lights. Such specialized terrestrial applications do not lend themselves readily to a generalized analysis. Therefore the purpose of this section is mainly to view the economics of SNAP for space power.

Although the costs associated with a single SNAP unit are relatively minor compared with the total cost of a space mission, several satellite systems are planned which may require several thousand electrical watts from many units. In such applications the cost of the power source becomes very significant.

Studies indicate that the energy from a 25- to 50-watt RTG fueled with ^{238}Pu, e.g., SNAP-9A, or with ^{242}Cm, e.g., SNAP-11, costs as much

as or more than energy from unoriented solar-cell systems of similar capacity. Figure III-1 shows that ^{242}Cm is apparently only competitive for short missions (1 year or less) and ^{238}Pu for long missions (5 years). A 25- to 50-watt RTG, fueled with ^{90}Sr at the 1964 anticipated cost of $1.50 per curie and separated from the payload by a boom rather than shielded, appears to be economically competitive with unoriented solar-cell generators of similar capacity suffering low radiation degradation during mission life and economically superior to unoriented solar-cell generators in orbits of high radiation damage. Comparison is not made with oriented solar-cell generators in the 25- to 50-watt range since they are generally employed at power levels above 100 watts unless the spacecraft itself is sun-oriented. Moreover, the supply of propellant for orienting purposes is limited to, at most, a few years.

The estimated relative advantage of 25- to 50-watt RTG's fueled with various isotopes that will or may be available in 1968 and thereafter is shown in Fig. III-2. The unit costs of isotopes in 1968 and thereafter used in this analysis are, at best, rough estimates and are subject to change as production alternatives are studied in greater detail. Again, strontium-fueled RTG's appear to offer the best potential for a space power system that is economically superior to solar cells. The cost of energy from an RTG fueled with ^{90}Sr costing 10 cents per curie is two to five times less than that from unoriented solar-cell generators. The cost of power from an RTG fueled with ^{244}Cm at $1000 per gram is shown to be roughly competitive with that from solar-cell generators, but, at $2000 per-gram, it is somewhat more costly than that from solar-cell generators and approximately a trade-off with that from a ^{238}Pu-fueled RTG. A thermionic generator with 10% overall efficiency fueled with ^{244}Cm at $1000 per gram might provide power at somewhat lower cost than an unoriented solar-cell system, or at about the same cost as one with ^{244}Cm at $2000 per gram.

The preceding data, presented on an energy cost basis, may also be expressed on a unit cost

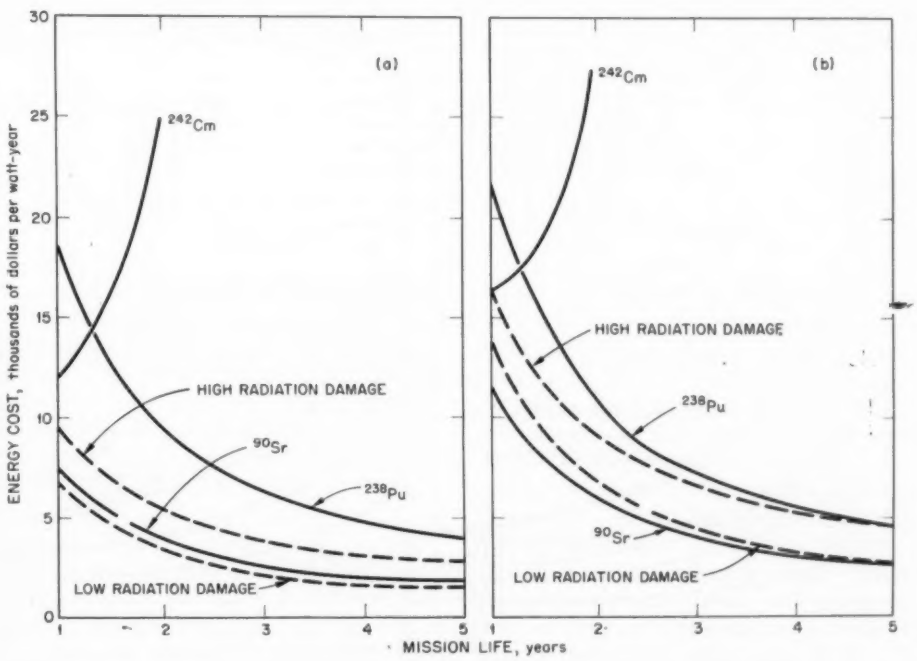

Fig. III-1 Cost of energy vs. mission life with 25- to 50-watt generator; 1964–1966 isotope costs: ^{90}Sr, $1.50 per curie; ^{238}Pu, $450 per gram; and ^{242}Cm, $12,700 per gram; ——, radioisotope SNAP generators. ----, unoriented solar cells. (*a*) $1000 per pound launching cost. (*b*) $5000 per pound launching cost.

basis. Table III-6 gives the calculated unit costs of a 25-watt RTG fueled with various isotopes at present and possible future costs. Comparison is made with a similar sized unoriented solar-cell generator. A launching cost of $5000 per pound was taken as representative of a small vehicle, e.g., Scout.

The calculated unit costs associated with a larger 150-watt RTG are given in Table III-7. Here comparison is made with a similar sized oriented solar-cell system; however, the weight of that system does not include the propellant necessary for orientation, and hence the launching cost is probably underestimated. Moreover, the assumed 3-year life probably exceeds the normal provision for orienting propellant. A launching cost of $3000 per pound was selected as more representative of a larger vehicle, e.g., Thor-Agena B or Atlas-Agena B.

Discussion

The justification of isotopic SNAP, as contrasted with alternative sources of power, is that such systems are superior in satisfying some or most of the following criteria:

1. Maximum reliability
2. Minimum system cost
3. Minimum space, weight, and payload interface requirements
4. Maximum useful life and radiation resistance
5. Maximum structural integrity
6. Compatibility with environment
7. Maximum safety and minimum radiation hazard

These criteria are, of course, of varying importance, depending upon the specific application.

The two 2.7-watt modified SNAP-3 and the two 25-watt SNAP-9A generators orbited to date have demonstrated the feasibility of isotopes in space, and they have provided valuable experience in the integration of these units into satellites and have established prelaunching interagency procedures and policies. A similar conclusion can be reached regarding the terrestrial applications to date. However, additional experience and quantitative evidence are required to judge more fully the merits of isotopic SNAP against the above criteria.

Major uncertainties related to the safety of large-scale use of isotopes in space, isotope availability, cost, sustained generator per-

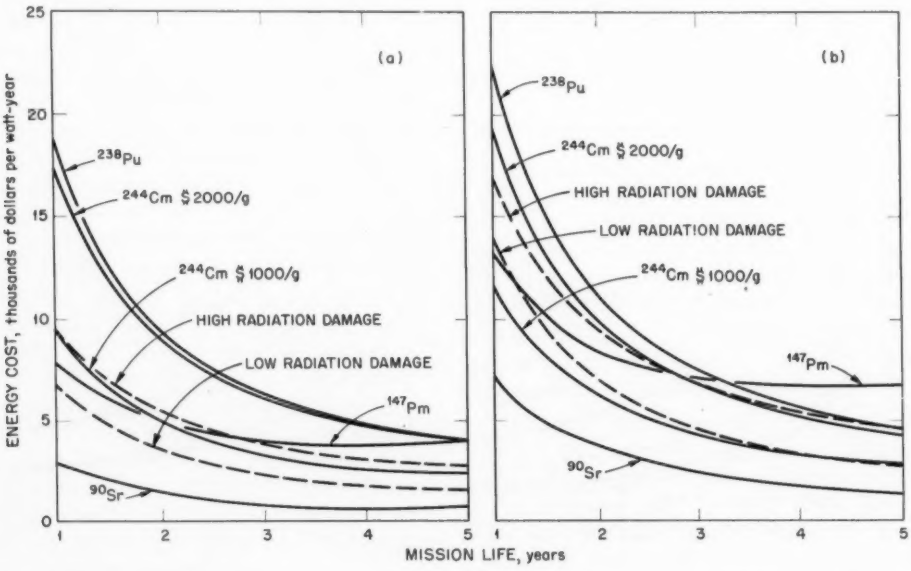

Fig. III-2 Cost of energy vs. mission life with 25- to 50-watt generator; assumed 1968–1972 isotope costs: ^{90}Sr, $0.10 per curie; ^{238}Pu, $450 per gram; ^{244}Cm, $1000 and $2000 per gram; and ^{147}Pm, $0.07 per curie. ——, radioisotope SNAP generators. ----, unoriented solar cells. (*a*) $1000 per pound launching cost. (*b*) $5000 per pound launching cost.

Table III-6 COST COMPARISON* OF 25-WATT RADIOISOTOPE THERMOELECTRIC GENERATOR AND UNORIENTED SOLAR-CELL GENERATORS

Isotope	Unit cost	Cost, thousands of dollars			
		Isotope	Generator	Launching	Total
^{244}Cm	$1000/g	200	35	65	300
	$2000/g	400	35	65	500
^{238}Pu	$450/g	410	35	100	545
^{90}Sr	$1.50/curie	130	35	125	290
	$0.10/curie	10	35	125	170
^{147}Pm	$0.07/curie	210	35	275	520
Unoriented solar cells					
Orbit of low radiation damage			125	210	335
Orbit of high radiation damage			230	250	480

*Assumptions:

1. Sufficient isotope and solar cells to provide 25 watts at end of 3-year mission, allowing for radioactive decay and solar-cell degradation.
2. Unit power:

^{244}Cm	2.8 thermal watts per gram of pure isotope
^{238}Pu	0.56 thermal watt per gram of pure isotope
^{90}Sr	0.90 thermal watt per gram of pure isotope
^{147}Pm	0.33 thermal watt per gram of pure isotope

3. 5% overall efficiency for isotopic thermoelectric generators.
4. Generator costs: For unoriented solar cells, $5000 per watt in orbits of low radiation damage; $9500 per watt in orbits of high radiation damage for 3 years; $1500 per watt for isotopic thermoelectric generators.
5. Generator weights:

^{244}Cm	0.5 lb/watt
^{238}Pu	0.8 lb/watt
^{90}Sr	1.0 lb/watt
^{147}Pm	2.2 lb/watt (3-year mission)
Solar cell	1.7 lb/watt assuming low radiation damage
	2.0 lb/watt allowing for high radiation damage

6. $5000 per pound launching cost.

formance, and ground handling of sources with high radiation backgrounds need to be resolved. A large-scale program for isotope heat-source application clearly depends on the resolution of these problems. The reluctance of NASA, DOD, and other agencies to establish firm requirements for isotopic space and terrestrial power systems is symptomatic of the above uncertainties.

The radiation hazard appears to be the key problem associated with future large-scale application of radioisotopes as heat sources in thermoelectric or thermionic generators. Isotopes can represent serious radiation hazards under certain types of system malfunctions. In space applications of isotopic SNAP, failure to achieve an intended orbit may result in the return of relatively undecayed isotopes either intact or as fallout, depending mainly upon the fuel capsule design. A thorough evaluation of the risks implicit in large-scale utilization of isotopes in space and guidance regarding the allowable magnitude of future applications are prerequisites to endorsing any large-scale program for using isotopes in space.

Terrestrial isotopic SNAP applications are of a more controllable nature, and it should be possible to design containment devices and to develop relatively insoluble chemical fuel forms that can meet safety standards. However, there is a need to provide guidance in system design first by developing safety standards.

Finally, the cost of the isotopic fuel can dominate the total cost of a SNAP device, as shown in Tables III-6 and III-7. Hence it should be a parameter that users factor into their

Table III-7 COST COMPARISON* OF 150-WATT RADIOISOTOPE THERMOELECTRIC GENERATOR AND ORIENTED SOLAR-CELL GENERATORS

Isotope	Unit cost	Cost, thousands of dollars			
		Isotope	Generator	Launching	Total
^{244}Cm	$1000/g	1200	150	110	1460
	$2000/g	2400	150	110	2660
^{238}Pu	$450/g	2470	150	225	2845
^{90}Sr	$1.50/curie	765	150	300	1215
	$0.10/curie	50	150	300	500
^{147}Pm	$0.07/curie	1255	150	585	1990
Oriented solar cells					
Orbit of low radiation damage			150	290	440
Orbit of high radiation damage			375	330	705

*Assumptions:

1. Sufficient isotope, solar cells, and propellant for solar-cell orientation, to provide 150 watts at end of 3-year mission, allowing for radioactive decay and solar-cell degradation.
2. Unit power:

^{244}Cm	2.8 thermal watts per gram of pure isotope
^{238}Pu	0.56 thermal watt per gram of pure isotope
^{90}Sr	0.90 thermal watt per gram of pure isotope
^{147}Pm	0.33 thermal watt per gram of pure isotope

3. 5% overall efficiency for isotopic thermoelectric generators.
4. Generator costs: For oriented solar cells, $1000 per watt in orbits of low radiation damage; $2500 per watt in orbits of high radiation damage for 3 years; $1000 per watt for isotopic thermoelectric generators.
5. Generator weights:

^{244}Cm	0.25 lb/watt
^{238}Pu	0.5 lb/watt
^{90}Sr	0.67 lb/watt
^{147}Pm	1.3 lb/watt (3-year mission)
Solar cell	0.65 lb/watt assuming low radiation damage
	0.73 lb/watt allowing for high radiation damage

6. $3000 per pound launching cost.

choice of a specific isotope as well as into their selection of a nuclear or nonnuclear system.

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Section

IV

Isotopes and Radiation Technology

Medicine

Production and Medical Uses of ^{67}Ga, ^{68}Ga, and ^{72}Ga

By J. J. Pinajian*

The possible usefulness of radioactive gallium in bone tumor detection and treatment was suggested by the preferential deposition of gallium in proliferating tissue.[1] This was discovered in toxicity and distribution studies[2–11] as a consequence of the consideration for use of gallium as a primary coolant in naval reactors.[12] After 3 years of animal studies, test doses were given to 12 patients with various types of bone malignancies. Lang,[13] in his published thesis, summarized the work of Dudley and included six case histories of the above 12 patients.

Medical Uses

The results of early studies with 14.2-hr ^{72}Ga were not very promising. Although the gallium isotope was highly concentrated in areas of tumor, there always appeared to be areas of low concentration; by the time the gallium was concentrated in the tumors, most of the radiation energy had been lost because of the short half-life; total-body irradiation was significant at subtherapeutic doses; and gallium toxicity appeared to limit repeated treatment. These conclusions were drawn from the extensive studies on the therapeutic use of gallium by the group at the Oak Ridge Institute of Nuclear Studies[14] and the U. S. Naval Hospital, National Naval Medical Center, Bethesda.[15] Twenty-one cases at Oak Ridge and four at Bethesda were given full therapeutic treatment. Only two cases, one at each hospital, showed X-ray evidence of bone repair. At Oak Ridge the tumor was a primary bone sarcoma, and at Bethesda it was carcinoma of the breast with bone metastasis. Varying degrees of subjective relief from deep bone pain were experienced in the treatment with ^{72}Ga at both places. It was felt that the severe bone-marrow depression following a full course of treatment was due to the radiation damage to the bone marrow and not to the effects of the stable gallium metal.[15]

A similar study[16] was made on four patients at the Brookhaven National Laboratory Research Hospital where the clinical findings were essentially those reported on by the Oak Ridge and Bethesda groups.

Carrier-free ^{66}Ga-^{67}Ga, produced in the Philips cyclotron at Amsterdam by the deuteron bombardment of zinc, was used by van der Werff[17] in a study of 15 patients, all of whom suffered from bone diseases. Some differences between the concentrations in pathological and normal bone were found, but the value of the general use of gallium as a therapeutic agent for bone tumors was considered doubtful. In a single case the influence of carrier was investigated, and no effect was found. No reference was made to the rather complete studies of the Bethesda group and the Oak Ridge group.

Recently, methods for gallium isotope preparation have been improved, and the possible use of 68-min ^{68}Ga, obtainable from a positron cow,[18,19] for external localization of brain tumors has been suggested,[20] along with ^{72}Ga for detecting osteoplastic changes in the skull.[21]

Preparation of Gallium Radioisotopes

Although 10 radioactive isotopes of gallium are known (Table IV-1), only the 77.9-hr ^{67}Ga, the 68-min ^{68}Ga, and the 14.2-hr ^{72}Ga are

*Oak Ridge National Laboratory.

Table IV-1 RADIOACTIVE ISOTOPES OF GALLIUM*

Isotope	Half-life	Electron capture	Radiations, Mev		
			β^-	β^+	γ
^{64}Ga	2.5 min			2.810 (38%)†	0.98
				6.090 (62%)	1.30
					2.25
					3.25
^{65}Ga	15.2 min			0.82 (10%)	0.053
				1.39 (19%)	0.118
				2.11 (56%)	1.38
				2.24 (15%)	1.86
^{65}Ga^m	8 min			(2.10 −)?	None
^{66}Ga	9.5 hr	ϵ		0.40 (0.7%)	0.83 (2)‡
				0.74 (1.3%)	1.04 (30)
				0.88 (4.7%)	1.37 (3)
				1.40 (~2%)	1.58 (?)
				1.75 (~0.5%)	1.93 (4)
				Others	2.18 (6)
					2.40 (2)
					2.75 (22)
					3.24 (2)
					3.41 (3)
					3.78 (2)
					4.12 (2)
					4.33 (5)
					4.83 (2)
^{67}Ga	77.9 hr	ϵ		< 0.01%	0.090 (2.7%)
					0.092 (63.9%)
					0.182 (29.6%)
					0.206 (1.07%)
					0.296 (20%)
					0.388 (4.9%)
					0.496 (0.4%)
					0.790 (0.2%)
					0.880 (0.4%)
^{68}Ga	68 min	$\epsilon/\beta^+ = 0.19$		0.800 (10%)	1.020
				1.900 (90%)	
^{70}Ga	21 min		1.65 (99.2%)		1.036
			0.61 (0.28%)		0.174
			0.44 (0.48%)		None
^{72}Ga	14.2 hr		0.64 (42%)		0.607 (7)
			0.96 (31%)		0.630 (21)
			1.51 (10%)		0.690 (~0)
			2.53 (9%)		0.768 (3)
			3.17 (8%)		0.810 (3)
					0.834 (94)
					0.894 (10)
					1.050 (7)
					1.317 (2)
					1.463 (4)
					1.595 (6)
					1.859 (6)
					2.203 (30)
					2.491 (10)
					2.508 (17)
					Others (< 1)
^{73}Ga	4.8 hr			1.19 (95%)	0.054 (9)
				0.40 (4.5%)	0.295 (97)
					0.745 (6)
					1.040 (1)
^{74}Ga	7.8 min			1.10	0.495 (7)
				2.00	0.600 (100)
				2.65	0.865 (8.5)
				2.7 (94)	1.02 (7)
				4.3 (6)	1.10 (5.5)
					1.20 (7.5)
					1.31 (2)
					1.45 (6.7)
					1.72 (5.5)
					1.84 (2.6)
					1.96 (6.7)
					2.35 (50)
					2.55 (2.5)
					2.72 (3.5)
					2.95 (3)
					Others (< 2)
^{75}Ga	2.0 min			3.3	0.58
^{76}Ga	32 sec			~6	0.57
					0.96
					1.12

*The decay-scheme information is taken from Ref. 26 and has been brought up to date where necessary (e.g., by use of Ref. 27, 28, 29, or 30).

†Intensities given in percentages refer to the number of unconverted photons emitted per 100 disintegrations.

‡Intensities given without the % symbol refer to the relative ratios of the unconverted photons.

available to people who do not have a reactor or accelerator.

GALLIUM-72

Gallium-72 with a minimum specific activity of 2000 mc per gram of gallium is offered by Oak Ridge National Laboratory (ORNL) at a price of $4.00 per millicurie in lots up to 500 mc and at $1.50 per millicurie in larger amounts. This radioisotope is made by the $^{71}Ga(n,\gamma)^{72}Ga$ reaction. A Ga_2O_3 target (100 mg of natural gallium) is irradiated for 61 hr in the pneumatic tube of the Low Intensity Test Reactor (LITR) at a flux of 10^{13} neutrons/(cm^2)(sec). The yield is 400 to 500 mc of ^{72}Ga, and the product has an ihitial specific activity of ~7 curies/g. Natural gallium is ~40% ^{71}Ga and 60% ^{69}Ga. Higher specific activity could be obtained with enriched targets, but they are too expensive to use.

Gallium for administration to human patients as the citrate is prepared[22] by dissolving the gallium oxide contained in the quartz ampule in concentrated hydrochloric acid with the gentle application of heat. The dissolved gallium is transferred with distilled-water rinses, indicator (bromcresol purple) and citric acid solutions are added, and sodium hydroxide is added to the end point (*p*H ~6). All the glassware, solutions, and materials are sterilized prior to use, and sterile techniques are used. For administration to the patient, a lead-shielded syringe may be used[23] or, alternatively, the material may be administered with an infusion apparatus,[14] using normal saline. The details of the standardization and counting techniques employed with the patient have been given.[24,25]

GALLIUM-68

Gallium-68 is usually obtained from a generator, or "cow,"[31] in which ^{68}Ga is formed in the decay of ^{68}Ge:

$$^{69}Ga(p,2n)^{68}Ge \xrightarrow[\text{e.c.}]{250\ \text{day}} {}^{68}Ga$$

The ^{68}Ga is made in the cyclotron from one of the following targets:

Target	Target form	Beam current, μa	Yield, mc/hr
Ga	Ni capsule	190–200	~0.4
Ga	Flat plate	500–700	~3
Ga_2O_3	Al capsule	190–200	~0.4

The ^{68}Ge is separated from the gallium by distilling $GeCl_4$, extracting the germanium from the distillate with CCl_4, and returning the activity to an aqueous phase. The aqueous solution is neutralized and evaporated, and the ^{68}Ge is recovered as magnesium orthogermanate, Mg_2GeO_4.

In a ^{68}Ga generator described by Gleason,[18] there is a solvent-extraction system using acetylacetone in cyclohexane as the organic phase. In a recently described ^{68}Ga generator,[19] ^{68}Ge is loaded onto alumina and is eluted, as needed, with 0.005*M* ethylenediaminetetraacetic acid adjusted to *p*H 7 with dilute NaOH. The operation of the ^{68}Ga generator is similar to that of other generators and may be described mathematically as follows:

The decay rate of radioisotope A is proportional to the number of atoms, N_A, present; i.e., at time t the activity of N_A is $\lambda_A N_A$. Then

$$\frac{dN_A}{dt} = -\lambda_A N_A \qquad (1)$$

At time $t = 0$, $N_A = N_{A_o}$; then

$$N_A = N_{A_o}\, e^{-\lambda_A t} \qquad (2)$$

The activity of the daughter B is $\lambda_B N_B$. The rate of change, dN_B/dt, in the number of N_B is equal to the growth of N_B from the decay of N_A, less the loss of N_B through decay:

$$\frac{dN_B}{dt} = \lambda_A N_A - \lambda_B N_B \qquad (3)$$

The number of daughter atoms, N_B, is given by

$$N_B = N_{A_o}\frac{\lambda_A}{\lambda_B - \lambda_A}\left(e^{-\lambda_A t} - e^{-\lambda_B t}\right) \qquad (4)$$

where N_{A_o} is the initial number of N_A atoms at $t = 0$. The decay rate of N_B is $\lambda_B N_B$, where

$$\lambda_B N_B = N_{A_o}\lambda_A \frac{\lambda_B}{\lambda_B - \lambda_A}\left(e^{-\lambda_A t} - e^{-\lambda_B t}\right) \qquad (5)$$

Since the decay rate of N_A at t is $\lambda_A N_{A_o} e^{-\lambda_A t}$,

$$\lambda_B N_B = N_A \lambda_A \frac{\lambda_B}{\lambda_B - \lambda_A}\left(1 - e^{-(\lambda_B - \lambda_A)t}\right) \qquad (6)$$

In a milking system or isotope generator where the half-life of the daughter is very much less

than that of the parent, i.e., $T_{\frac{1}{2}}(A) \gg T_{\frac{1}{2}}(B)$, then $\lambda_B \gg \lambda_A$. Equation 6 then reduces to

$$\lambda_B N_B = \lambda_A N_A \ (1 - e^{-\lambda_B t}) \qquad (7)$$

GALLIUM-67

Carrier-free 77.9-hr ^{67}Ga has been produced as a secondary product in the irradiation of enriched ^{68}ZnO by the ^{68}Zn$(p,2n)^{67}$Ga reaction.[32] The primary product, ^{67}Cu, was, after dissolution of the target in hydrochloric acid, removed by ion-exchange chromatography. The ^{67}Ga was extracted with isopropyl ether from the hydrochloric acid solution. Approximately 30 mc/(ma)(hr) was obtained from the irradiation of 62 mg of ^{68}ZnO in a window target,[33] a 70-μa beam of 23-Mev protons being used. Other possible reactions for producing ^{67}Ga are:

$$^{67}\text{Zn}(p,n)^{67}\text{Ga}$$
$$^{66}\text{Zn}(d,n)^{67}\text{Ga}$$
$$^{65}\text{Cu}(\alpha,2n)^{67}\text{Ga}$$

Clinical Uses of ^{125}I

In an earlier issue of *Isotopes and Radiation Technology*, methods for making ^{125}I and its uses were reviewed, and X-ray photographs taken with it were shown.[34] The increasing importance of this isotope in the medical field is indicated by information presented at a symposium[35] in Chicago earlier this year, sponsored by the Volk Radiochemical Company in cooperation with the Central Chapter of the Society of Nuclear Medicine. The physical and chemical aspects of this isotope, as well as biomedical research and clinical applications, were discussed.

Because of its nearly monochromatic 0.0274-Mev photons, ^{125}I is finding application as an X-ray source as well as as a tracer. The good monochromaticity and constant intensity of its emitted beam were mentioned by Cameron as making it superior to conventional X-ray sources in this application.

Beck described the scintillation detectors used for the K X rays and 35.4-Mev gamma rays in organ scanning as having a focused collimator with very thin septa to give good resolution and thin crystals to give a high photopeak crystal efficiency. Production of ^{125}I from ^{124}Xe in natural xenon was described by Erlebach of Canada and from xenon enriched in ^{124}Xe by Baker of ORNL. The lower radiation dose to a patient from ^{125}I scanning than from ^{131}I was shown by Harper and Lipscomb from Argonne Cancer Research Hospital. Clayton et al. described a photographic method of thyroid visualizing that would eliminate the need for elaborate scanning equipment. In this method two films are exposed simultaneously, or Polaroid film is used with a thin thallium-activated sodium iodide layer.

Because of the lower radiation dose to the patient, ^{125}I is becoming important in thyroid scanning studies, as discussed by Charkes. Other clinical uses include labeling of albumin for blood volume studies (Williams) and for protein transfer-rate determinations (Bruce and Mount), of diiodofluorescein in diagnosing intraocular tumors (Newell), and of Hippuran for urological diagnoses (Winter). Bacteria labeled with ^{125}I were reported (Harford) to give as good autoradiographs as tritium, which is now used.

Iodine-125 has also been used in conjunction with ^{131}I for double-labeling experiments. For example, antitumor antibodies are localized by labeling the antiprotein with one isotope and normal protein with the other, injecting the mixture into a tumor-bearing animal, and determining the amounts of each in various organs (Yagi and Pressman). For metabolic studies a precursor and its end product can be similarly labeled and followed (Levy).

The general program chairman was Paul V. Harper, M.D., Department of Surgery, Argonne Cancer Research Hospital, The University of Chicago, and session chairmen included Wm. G. Myers, Ph.D., M.D., University Hospital, Columbus, Ohio, and Lester M. Levy, M.D., Long Island Jewish Hospital, New Hyde Park, N. Y. The program included abstracts of the talks listed on the following page.

Physical and Chemical Aspects of ^{125}I

Instrumentation for the Detection of ^{125}I
Robert N. Beck, B.S.
Argonne Cancer Research Hospital, Chicago, Ill.

The Radiochemistry and Production of ^{125}I
W. E. Erlebach
Atomic Energy of Canada, Ottawa, Canada

^{125}I Production Studies at Oak Ridge National Laboratory
P. S. Baker, Ph. D.
Oak Ridge National Laboratory, Oak Ridge, Tenn.

The Assay and Dosimetry of ^{125}I
Paul V. Harper, M.D., and Charles Lipscomb, M.D.
Argonne Cancer Research Hospital, Chicago, Ill.

^{125}I as a Source of X Rays
John R. Cameron, Ph. D.
University of Wisconsin, Madison, Wis.

Application of ^{125}I to Direct Photographic Visualization of the Thyroid
Glenn Clayton, B.S., Joseph Kluski, Theodore Fields, M.S., Ervin Kaplan, M.D.
Veterans Administration Hospital, Hines, Ill.

Biomedical Research Applications of ^{125}I

Pair-Labeled Antitumor Antibodies
Yasuo Yagi, Ph. D., and David Pressman, Ph. D.
Roswell Park Memorial Institute, Buffalo, N. Y.

Electron Microscopic Autoradiography of Bacteria Labeled with ^{125}I
Carl G. Harford, M.D.
Washington University School of Medicine, St. Louis, Mo.

Use of ^{125}I in Double-Isotope-Labeling Techniques for Metabolic Studies
Lester M. Levy, M.D.
Long Island Jewish Hospital, New Hyde Park, N. Y.

Use of ^{125}I for Determination of Transfer Rates of Proteins
W. R. Bruce, Ph. D., and D. Mount, M.D.
Ontario Cancer Institute, Toronto, Canada

Clinical Applications of ^{125}I

Utilization of ^{125}I for Thyroid Scanning
N. David Charkes, M.D.
Albert Einstein Medical Center, Philadelphia, Pa.

Clinical Application of ^{125}I-Labeled Albumin
John A. Williams, M.D.
Harvard Medical School–Beth Israel Hospital, Boston, Mass.

^{125}I-Labeled Diiodofluorescein in the Diagnosis of Intraocular Tumor
Frank W. Newell, M.D.
Department of Surgery (Ophthalmology), University of Chicago, Chicago, Ill.

Use of ^{125}I in Diagnostic Urology
Chester C. Winter, M.D.
Ohio State University Hospitals, Columbus, Ohio

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Production and Medical Uses of ^{67}Ga, ^{68}Ga, and ^{72}Ga

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Section

V

Isotopes and Radiation Technology

Miscellaneous Items of Interest

Conferences

200 ATTEND RADIOISOTOPE CONFERENCE ON INDUSTRIAL, BIOMEDICAL APPLICATIONS

Some 200 representatives of industrial, university, and private research laboratories met in Gatlinburg, Tenn., April 19–22 for the Second Annual Oak Ridge Radioisotope Conference. The Conference was sponsored by Oak Ridge Institute of Nuclear Studies (ORINS) and Oak Ridge National Laboratory (ORNL) in cooperation with the U. S. Atomic Energy Commission (AEC). A number of industries exhibited equipment for use in radioisotope work. One feature that attracted considerable attention was the ORINS mobile training laboratory, which is taken to college campuses for use in teaching techniques of handling radioisotopes. The unit has laboratory facilities for 8 to 10 students.

Conference proceedings, which included papers on industrial, medical, and biological uses of isotopes, will not be published, but long abstracts, including many tables and illustrations, were preprinted in Second Annual Oak Ridge Radioisotope Conference (USAEC Report TID-7689). The program was:

Diagnostic Uses for $^{99}Tc^{m}$	Katherine A. Lathrop and Paul V. Harper, University of Chicago
Rapid Screening Method for Detecting Abnormal Plasma Vitamin B_{12} Binding Sites in Chronic Myelogenous Leukemia Using Arm Counting	C. C. Lushbaugh and William Gibbs, ORINS
Evaluation of ^{125}I Measurement Techniques for Clinical Applications of Radioiodine	James S. Eldridge and Peter Crowther, ORNL
Use of ^{125}I as a Label in Immunologic Studies	Frank W. Fitch, Katti Dzoga, Howard Benensohn, and Paul V. Harper, University of Chicago
Comparative Study of Thyroid and Whole Body Retention of Iodine in Box Turtles	W. Gibbs, E. Wilson, H. Hodges, and C. C. Lushbaugh, ORINS
Thyroidal and Extrathyroidal Hormone Metabolism in Children	Leo Oliner, Veterans Administration Hospital and Indiana University School of Medicine
Comparison of Digital and Analog Methods for Radioisotope Measurement	Bergene Kawin, Frank V. Huston, Veterans Administration Hospital, Fort Howard, Md.
Isotopes Information Center at ORNL—A Progress Report	Robert H. Lafferty, Jr., Martha Gerrard, P. S. Baker, and D. A. Fuccillo, ORNL
Potentials of Isotopic Radiography	M. L. Pool and Harry D. Spangenberg, Jr., Ohio State University
Light Filter for Reduction of Background in Liquid Scintillation Counting	Howard A. Swartz, Butler University
Maximum Allowable Radiotracer Injections for Flow Rate Measurements in Public Streams	R. P. Gardner, Research Triangle Institute

Calibration of Radiation Monitors for Process Waste Water	R. L. Shipp, ORNL
Some Uses of Fluid Tracers in Oil Wells	R. George Mihram, Halliburton Company, Duncan, Okla.
Synthesis, Storage, and Manipulation of Tritiated Steroid Estrogens of High Specific Activity	H. I. Jacobson, N. N. Saha, H. G. Neumann, V. Colucci, and E. V. Jensen, University of Chicago
Use of ^{35}S Acetazolamide and Other Labeled Drugs in the Study of Blood Brain Barrier Changes	V. Nair and L. J. Roth, University of Chicago
Use of Isotopes To Establish a Method for Determining the Permeability Characteristics of the Pulmonary Membrane	Aubrey E. Taylor, Arthur C. Guyton, Vernon S. Bishop, and Kirby B. DeLozier, University Medical Center, Jackson, Miss.
Biological Behavior of ^{137}Cs in Relation to That of Potassium	B. V. Rama Sastry, Vanderbilt University School of Medicine
Complications in Residue Studies with Radioisotopic Drugs	Charles Rosenblum, Dominick J. Tocco, and Eugene E. Howe, Merck Sharp & Dohme Research Laboratories
Interspecific Transfer of ^{45}Ca and ^{32}P by Root Systems	Frank W. Woods and Kirby Brock, Duke University and U. S. Forest Service, Aiken, S. C.
Mechanisms of Ion Uptake by Leaves of Higher Plants as Revealed by Radioisotopes	S. H. Wittwer, Y. Yamada, W. H. Jyung, and M. J. Bukovac, Michigan State University
Using $^{14}CO_2$ for Carbon Fixation Studies with Pollen	James A. Goss, Kansas State University
Use of Radioisotopes in Marine Biological Research	Thomas W. Duke, Bureau of Commercial Fisheries, Beaufort, N. C.
Radioisotope Powered Underwater Acoustic Beacon	Marvin P. Norin, Martin Company
Using Gamma Transmission To Determine the Shielding Effectiveness of Space Vehicles	A. D. Krumbein and P. S. Mittelman, United Nuclear Corp.
Report on Compilation of Some Beta-Excited Spectra	L. E. Preuss, H. Collins, and E. R. Wilson, Edsel B. Ford Institute for Medical Research
Radioisotopes in Alkaline-Earth Analysis	S. A. Reynolds, ORNL
Mechanism of Methylene Blue Dye Adsorption on Siliceous Minerals	C. S. Brooks, Shell Development Company, Houston, Tex.
Surface Self-Diffusion of Adsorbed Atoms	H J. Arnikar, E. A. Daniels, and O. P. Mehta, University of Poona, India
Recycle Gas Chromatography: Separation of Isotopic Molecules	John W. Root, Edward K. C. Lee, and F. S. Rowland, University of Kansas
Use of ^{35}S for Investigation of Isotopic Exchange Reactions of Elemental Sulfur	Charles W. Owens and F. S. Rowland, University of New Hampshire and University of Kansas
Use of Isotopes in Ring and Linear Vacuum Distillation Source Studies	C. Bugenis and Luther E. Preuss, Edsel B. Ford Institute for Medical Research
Unique Scintillator for Neutron Detection	H. H. Ross and R. E. Yerick, ORNL
How To Obtain More Fracturing Information by the Use of Radioisotopes	R. George Mihram, Halliburton Company, Duncan, Okla.
Study of the Sources of Ceramic Inclusions in Cast Gas Turbine Components by Radiotracer Analysis	L. L. Packer, J. N. Williams, and C. M. Phipps, United Aircraft Corp. Research Laboratories and Pratt & Whitney Division of United Aircraft Corp., East Hartford, Conn.

Frequency Response Testing of Aqueous Mixing Systems by a Radiotracer Pulse Technique	R. P. Gardner, L. F. Ballard, and J. K. Ferrell, North Carolina State College
Acid Circulation, Volume, Replacement, and Entrainment Measured in an Alkylation Plant with Radiotracer	D. E. Hull, B. A. Fries, and J. T. Gilmore, California Research Corp., Richmond, Calif.
Residence Time of Particles in a Rotary Kiln	B. A. Fries, California Research Corp., Richmond, Calif.
Tracer Studies of Impurity-Doped Oxide Cathodes	James K. Lee, Television Picture Tube Division, Radio Corp. of America
Wear Measurement of Machine Parts by Activation Analysis	Henry L. Wood, Virginia Polytechnic Institute, and Benjamin A. A. Logerquist, U. S. Army
Sediment Gauge for Rivers and Streams	J. Papadopoulos, F. B. Sellers, and C. A. Ziegler, Parametrics, Inc., Waltham, Mass., and J. R. McHenry, USDA Sedimentation Laboratory, Oxford, Miss.

AEC Activities

LIST OF RADIOISOTOPE LICENSEES AVAILABLE

A list of 6000 names of radioisotope licensees is now available at cost from the AEC. This information, known as *List D, Byproduct Material (Radioisotopes)*, is coded to identify the kinds of organizations holding the licenses. Three other lists are available: A, Facilities; B, Special Nuclear Material; and C, Source Material.

Copies of lists may be purchased at a production cost of $4.60 per hour of Addressograph machine time. The average time to run list D is 4 hr. Orders for reproduction of lists or requests for further details should be addressed in writing to the U. S. Atomic Energy Commission, Office of the Secretary, Public Proceedings Branch, Washington, D. C., 20545.

AEC CHANGES REGULATORY ORGANIZATION; RAISES LICENSING, STANDARDS DEVELOPMENT TO RANK OF DIVISIONAL FUNCTIONS

AEC has announced organizational changes in its regulatory program. The new organization reflects the growth of the atomic energy industry and the increase in AEC regulatory activities by according divisional status to both the licensing and the standards-development functions of the regulatory staff.

In March 1961 the Commission separated its regulatory functions from its operational and developmental functions at the staff level by establishing the position of Director of Regulation. Harold L. Price, the Director of Regulation, reports directly to the Commission on the work of the three divisions that heretofore have carried out the regulatory staff work of the Commission—the Division of Licensing and Regulation, the Division of Radiation Protection Standards, and the Division of Compliance.

In the new organizational changes, the functions of the Division of Licensing and Regulation and of the Division of Radiation Protection Standards are assigned to four new divisions: the Division of Materials Licensing, the Division of Reactor Licensing, the Division of Safety Standards, and the Division of State and Licensee Relations. The organization of the Division of Compliance remains the same. All five divisions will report to Price.

The Division of Materials Licensing will license the uses of radioisotopes, the atomic energy source materials uranium and thorium, and the special nuclear materials ^{233}U, ^{235}U, and Pu.

The new Division of Reactor Licensing will handle the staff review of applications for licenses to build and operate nuclear reactors; licensing of reactor operators; evaluation of nuclear safety aspects of Commission-owned and Department of Defense-owned reactors that are exempt from licensing; and liaison with the Advisory Committee on Reactor Safeguards.

The Division of Safety Standards will develop and recommend nuclear safety standards to

protect employees and the public; work on standards for the design, location, construction, and operation of reactors and other nuclear facilities; provide technical advice to federal and state agencies and other AEC divisions; provide staff assistance to the Commission in matters involving the Federal Radiation Council; and participate in nuclear safety research programs.

The Commission's program of cooperation with the states leading to agreements for the transfer to the states of part of AEC's regulatory authority will be carried out by the Division of State and Licensee Relations. It also will handle such activities with AEC licensees as indemnification, enforcement, and the licensing for export of radioactive materials and nuclear facilities.

The new organizational chart of the regulatory staff is shown in Fig. V-1.

AEC SIMPLIFIES LICENSING REGULATIONS FOR PLUTONIUM, AMERICIUM SOURCES

AEC has simplified the licensing of small quantities of all plutonium isotopes and ^{241}Am in sources used for calibration of nuclear instruments and as references.

The amendments to Commission regulations, Part 70 ("Special Nuclear Material") and Part 30 ("Licensing of Byproduct Material"), are substantially the same as those published as proposed rules in the *Federal Register* on Sept. 20, 1963. The amendments became effective in June.

Under the amendments, general licenses are issued for up to 5 μc of these isotopes provided they have been manufactured by a person or firm holding a specific license from either the AEC or a state that has an agreement with AEC for the regulatory control of certain radioactive materials. A general license requires no application to the Commission. Requirements are established for specific licenses for persons who manufacture sources for distribution.

The amendment to Part 30 also includes a general license for the export of ^{241}Am to foreign countries other than Sino-Soviet bloc countries.

FOUR APPOINTED TO AEC COMMITTEE ON MEDICAL APPLICATIONS OF ISOTOPES

Chairman Glenn T. Seaborg has appointed four additional members to the AEC's Advisory Committee on Medical Uses of Isotopes. The new members are E. Richard King, M.D., Professor of Radiology, Medical College of Virginia; Robert H. Greenlaw, M.D., Associate Professor of Radiology, University of Kentucky; Lee Edward Farr, M.D., Professor of Nuclear and Environmental Medicine and Chief, Section of Nuclear Medicine, M. D. Anderson Hospital and Tumor Institute, University of Texas; and Harald H. Rossi, Ph. D., Professor of Radiology, College of Physicians and Surgeons, Columbia University.

There are seven other members who will continue to serve on the Committee. It was explained that the volume of work has increased as a result of the widespread uses of radioisotopes in medicine.

The Committee, established in 1958, advises the Commission and the AEC Director of Regulation on matters relating to policies and standards for the regulation and licensing of medical uses of radioisotopes in humans. Present members are: Wallace D. Armstrong, M.D., University of Minnesota; Reynold F. Brown, M.D., University of California and New York Life Insurance Company; Donald S. Childs, M.D., Mayo Clinic; John A. D. Cooper, M.D., Ph.D., Northwestern University Medical School; George V. LeRoy, M.D., University of Chicago; Edith H. Quimby, Sc.D., Columbia University; and Rulon W. Rawson, M.D., Memorial Hospital, Cornell University Medical College, and Sloan-Kettering Institute for Cancer Research, New York.

AEC APPROVES EXPORT OF RADIOISOTOPES FOR WORK AT CZECH, HUNGARIAN INSTITUTES

AEC has approved the export of two shipments of radioactive materials to Czechoslovakia and one to Hungary for use in research.

New England Nuclear Corporation of Boston was granted a license to export 3.7 mc of *D*-glucosamine-1-^{14}C hydrochloride to OMNIPOL Foreign Trade Corporation for use at the Institute for Research on Rheumatic Illnesses in Prague.

Schwarz BioResearch, Inc., Orangeburg, N.Y., was issued a license for export of 250 μc of tritium-labeled uridine and 250 μc of tritium-labeled histidine for biochemical research by the Biophysical Institute of the Czechoslovakia Academy of Sciences in Brno.

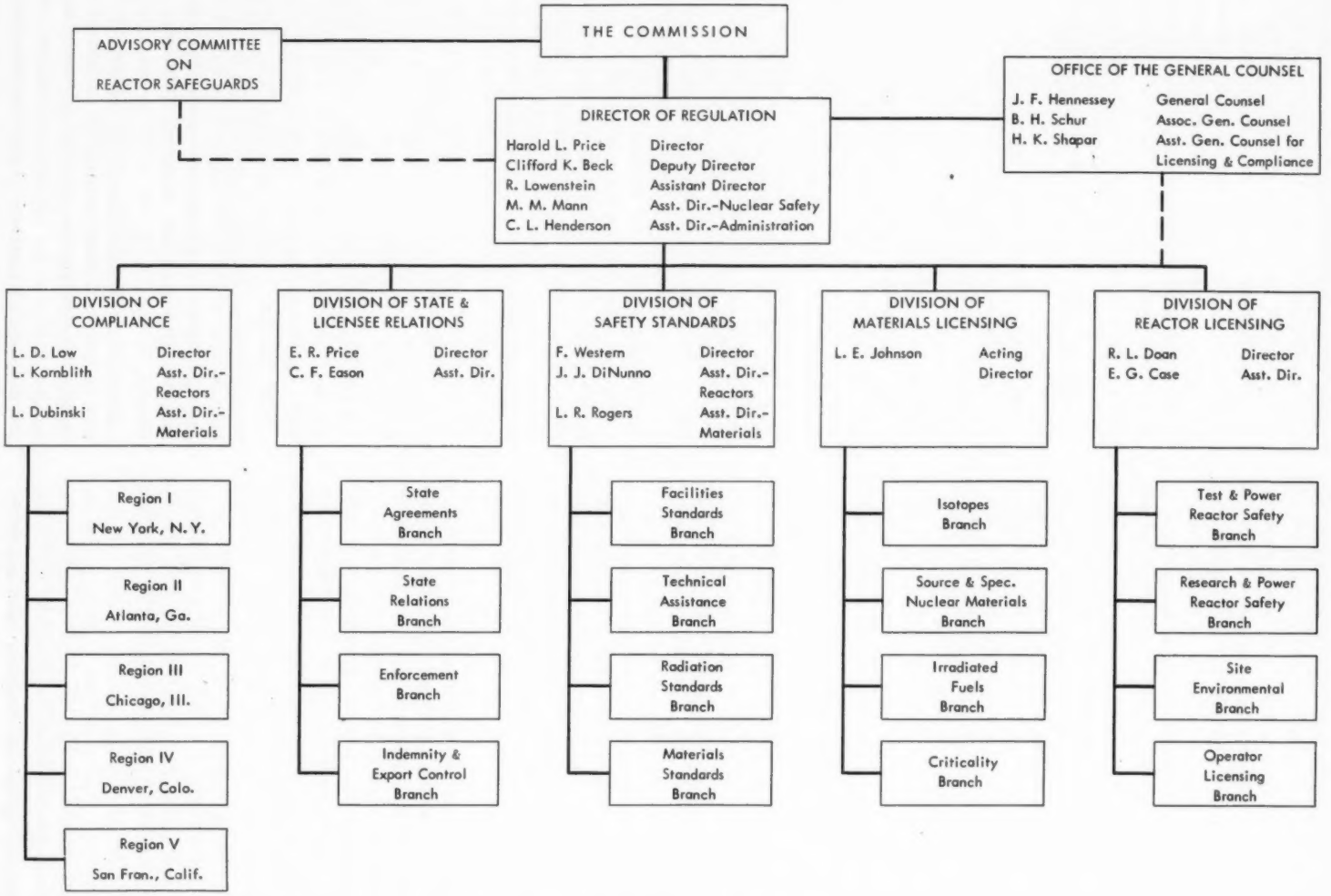
THE COMMISSION
ADVISORY COMMITTEE ON REACTOR SAFEGUARDS
DIRECTOR OF REGULATION
Harold L. Price — Director
Clifford K. Beck — Deputy Director
R. Lowenstein — Assistant Director
M. M. Mann — Asst. Dir.-Nuclear Safety
C. L. Henderson — Asst. Dir.-Administration
OFFICE OF THE GENERAL COUNSEL
J. F. Hennessey — General Counsel
B. H. Schur — Assoc. Gen. Counsel
H. K. Shapar — Asst. Gen. Counsel for Licensing & Compliance
DIVISION OF COMPLIANCE
L. D. Low — Director
L. Kornblith — Asst. Dir.-Reactors
L. Dubinski — Asst. Dir.-Materials
DIVISION OF STATE & LICENSEE RELATIONS
E. R. Price — Director
C. F. Eason — Asst. Dir.
DIVISION OF SAFETY STANDARDS
F. Western — Director
J. J. DiNunno — Asst. Dir.-Reactors
L. R. Rogers — Asst. Dir.-Materials
DIVISION OF MATERIALS LICENSING
L. E. Johnson — Acting Director
DIVISION OF REACTOR LICENSING
R. L. Doan — Director
E. G. Case — Asst. Dir.
Region I New York, N. Y.
Region II Atlanta, Ga.
Region III Chicago, Ill.
Region IV Denver, Colo.
Region V San Fran., Calif.
State Agreements Branch
State Relations Branch
Enforcement Branch
Indemnity & Export Control Branch
Facilities Standards Branch
Technical Assistance Branch
Radiation Standards Branch
Materials Standards Branch
Isotopes Branch
Source & Spec. Nuclear Materials Branch
Irradiated Fuels Branch
Criticality Branch
Test & Power Reactor Safety Branch
Research & Power Reactor Safety Branch
Site Environmental Branch
Operator Licensing Branch

Fig. V-1 AEC regulatory organization.

The Agricultural Research Service of the U. S. Department of Agriculture will export fertilizer containing 825 mc of ^{32}P to the Research Institute for Irrigation and Rice Cultivation, Szarvas, Hungary. Field experiments there with the tracer include determination of the most efficient methods of rice fertilization. The work is a continuation of experiments being sponsored by the International Atomic Energy Agency, of which Hungary is a member nation. AEC previously authorized shipments of 175 and 360 mc of ^{32}P-labeled fertilizer to Hungary for the studies.

In authorizing these shipments, AEC noted that the material to be exported has no strategic or military value.

Education

REGULATION, INDUSTRY, RADIATION, AND NEUTRON-ACTIVATION FILMS AVAILABLE

The following new color movies are available from AEC film libraries: "Regulation of Atomic Radiation," "Radioisotopes — Safe Servants of Industry," "Man and Radiation," and "Neutron Activation Analysis." The first three of these are popular-level, and the fourth, professional-level, 16-mm films varying in length from approximately 30 to 40 min.

The first film surveys the regulation of sources and uses of radiation; AEC licensing procedures; the setting up of radiation-protection standards; safe handling from production to packaging, shipping, and use; and waste disposal.

The second film describes gaging, radiography, tracing, and other aspects of radioisotopes as safe industrial tools. It shows various applications and practices in the use of these materials in industry today.

The third film covers irradiation of wood, plastics, and food, as well as other applications, including agriculture, medicine, industry, power, and research.

The fourth film describes the potential applications of neutron-activation analysis and shows the kinds of neutron sources used (isotopic, accelerator, and nuclear reactor) and the latest counting techniques employed (especially those of multichannel gamma-ray spectrometry and spectrum stripping). Examples of recent applications in the fields of scientific crime detection, geology and geochemistry, agriculture, medicine, the petroleum and chemical industries, and the semiconductor industry are shown.

These may be borrowed without charge from the AEC's domestic libraries as well as from their offices in Tokyo, Brussels, Buenos Aires, and London, and from the International Atomic Energy Agency office in Vienna.

ACTIVATION ANALYSIS COURSES TO BEGIN

The Activation Analysis Group of General Atomic plans to offer a number of specialized 1-week courses and a longer, more general, 2-week course on neutron-activation analysis.

The first of these courses will be on determination of oxygen in metals by means of activation analysis with 14-Mev neutrons produced by small moderate-cost accelerators. It will be held in General Atomic's San Diego laboratories August 3–7, 1964.

The oxygen-in-metals course will be a full-time course, 40 hr, and will include both extensive lecture coverage and laboratory work. To ensure maximum effectiveness of the laboratory work, the course will be limited to 12 students.

For additional information on this course and projected later ones, write: R. M. Watkins, Manager, Applications Development, Activation Analysis Service, General Dynamics/General Atomic, P. O. Box 608, San Diego, Calif., 92112.

CALCIUM-47 BROCHURE

A pamphlet entitled *Production of Calcium-47, with Literature Review of Uses* has been written by the staff of the Isotopes Information Center at ORNL. It describes production techniques, lists a few of the uses of the isotope, and includes approximately 80 references.

Copies may be obtained free of charge from the Division of Technical Information Extension, USAEC, P. O. Box E, Oak Ridge, Tenn., 37831, by asking for ORNL-IIC-2.

ORINS OFFERS GEOCHEMISTRY COURSE

A laboratory-lecture course on "Nuclear Methods in Geochemistry" will be presented October 6–20, 1964, at the Special Training Division of the Oak Ridge Institute of Nuclear Studies.

Topics to be covered include trace-element and gross-compound determinations by activa-

Fig. V-1 AEC regulatory organization.

tion analysis with slow and fast neutrons in geological samples (rocks, minerals, and petroleum) and absolute dating by different techniques. Geological problems, including sediment transport, soil and beach erosion, and water balance, will also be discussed in the light of new techniques and developments.

Geologists and other scientists interested in these and related problems can obtain further information about the course from Dr. Ralph T. Overman, Chairman, Special Training Division, Oak Ridge Institute of Nuclear Studies, P. O. Box 117, Oak Ridge, Tenn., 37831.

Availability of Isotopes and Services

NEW ISOTOPE INDEX *EDITION PUBLISHED*

The seventh edition of *The Isotope Index*, edited by J. L. Sommerville, has recently been published. This book, which provides an up-to-date listing of the availability and price of radioisotopes, stable isotopes, and labeled compounds, may be bought for $7.00 for paperback or $8.00 for hardbound copy from Scientific Equipment Company, P. O. Box 19086, Indianapolis 19, Ind. Quantity discounts are available.

FIRM OFFERS RADIONUCLIDE SELECTION

The Special Sources Department of New England Nuclear Corporation has undertaken a program of providing a comprehensive selection of both accelerator-produced and reactor-produced radionuclides. Gamma spectra and complete specifications accompany each shipment. The following isotopes are available from stock:

^{207}Bi (carrier free)	^{54}Mn (carrier free)
^{57}Co (6 mc/μg Co)	^{22}Na (2 mc/mg Na)
^{195}Au (carrier free)	^{85}Sr (carrier free)
^{55}Fe (carrier free)	^{119}Sn^{m} (>25 mc/g)

The following carrier-free isotopes are in process and should be available as noted:

^{7}Be (July)	^{125}I (September)
^{109}Cd (July)	^{44}Ti(^{44}Sc) (June)
^{58}Co (May)	

ORNL INVENTORIES HIGH-PURITY ^{66}Zn

Several grams of 99.35% ^{66}Zn are available for sale from Isotopes Sales Department, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, Tenn., 37831.

General

IMPACT OF FOOD IRRADIATION STUDIED

The Business and Defense Services Administration of the U. S. Department of Commerce has recently set up a five-man staff, headed by J. M. Schaffer, to study industrial and consumer effects that may result from the radiation preservation of food. This group will study various aspects of the irradiated food program such as the increase in U. S. food exports that could result from the use of radiation preservation techniques, the potential use of irradiated food to provide proteins to developing areas that do not have conventional food-storage facilities, and the impact of irradiation on other newer methods of food processing such as freeze-drying.

ACTIVATION ANALYSIS BRINGS FIRST CONVICTION

The AEC has been advised that evidence developed through the use of activation analysis has been used for the first time in a criminal trial in the United States. Attorney General Robert F. Kennedy said that use of the technique helped to establish, to the satisfaction of judge and jury, that illicit whiskey seized on a truck in one state had been transported from a still in a distant state.

Samples of soil were scraped from the bottom of a tractor-trailer truck seized in New York; later, soil samples were collected from the site of the suspected still in Georgia. These samples were activated at the Armed Forces Radiobiology Research Institute, Bethesda, Md., for the Internal Revenue Service, and the analytical results showed that the mud samples taken in New York matched those obtained in Georgia. [For a review of activation analysis, see the Fall 1963 issue of *Isotopes and Radiation Technology*, 1(1): 64-77.]

UNION CARBIDE AWARDS SUBCONTRACT FOR HIGH FLUX ISOTOPE REACTOR FUEL

A subcontract has been awarded to Metals and Controls, Inc., Attleboro, Mass., for furnishing complex nuclear reactor fuel assemblies for the High Flux Isotope Reactor (HFIR). Each assembly, although only about the size of an ordinary wastebasket, will be a complete fuel core and will be capable of generating about 100 Mw of heat.

The award by Union Carbide Corporation, Nuclear Division, which operates the Oak Ridge National Laboratory for the Atomic Energy Commission, was on a cost-plus-fixed-fee basis, with the total amount estimated at $1,851,233. Thirty-three fuel assemblies, sufficient to operate the HFIR for 18 months, will be furnished. Fuel in the HFIR is expected to be replaced every 14 to 17 days under routine full-power operating conditions.

The HFIR, which will have the highest neutron flux of any reactor in the United States, 5×10^{15} neutrons/(cm^2)(sec), will be used to produce research quantities of transuranium isotopes—principally americium, curium, and californium. The objective of the AEC transuranium program is production of these elements in milligram to gram amounts to permit extension of heavy-element research. A description of the HFIR was published recently by F. T. Binford and E. N. Cramer (Eds.), The High Flux Isotope Reactor, A Functional Description, Volume 1, USAEC Report ORNL-3572, Oak Ridge National Laboratory, May 1964.

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